IONS-04 (INTEX Ozonesonde Network Study, 2004): Perspective on Summertime UT/LS (Upper Troposphere/Lower Stratosphere) Ozone over Northeastern North America

24 April 2006

Submitted to *J. of Geophysical Research - Atmospheres* (INTEX Issue) 2006JD007441

Anne M Thompson[1], Jesse B Stone[1], Jacquelyn C Witte[2], R Bradley Pierce[3], Robert B Chatfield[4], Samuel J Oltmans[5], Owen R Cooper,[6] Brett F Taubman[1], Bryan J Johnson[5], Everette Joseph[7], Tom L Kucsera[2], John T Merrill[8], Gary A Morris[9], Scott Hersey[10], Michael J Newchurch[11], F J Schmidlin[12], David W Tarasick[13], Valerie Thouret[14], Jean-Pierre Cammas[14]

[1] Meteorology Department, Penn State University, 503 Walker Bldg, University Park, PA 16802-5013; 814-865-0479, fax-814-865-3663; anne@met.psu.edu

[2] SSAI (Lanham, MD 20706); also at NASA/Goddard Space Flight Center/Code 613.3, Greenbelt, MD 20771; 301-614-6046, 301-614-5903; witte@gavial.gsfc.nasa.gov; tlk@croc.gsfc.nasa.gov

[3] NASA/Langley Research Center, Atmospheric Sciences Competency, Hampton, VA 23681; 757-864-5918; fax, 757-864-7790; r.b.pierce@larc.nasa.gov

[4] NASA/Ames Research Center, SGG Division, Moffett Field, CA 94035; 650-604-5490; fax 650-604-3625; chatfield@clio,arc.nasa.gov

[5] NOAA/Earth Systems Research Laboratory, Boulder, CO 80305; 303-497-6676 (SJO); 303-497-6842 (BJJ); samuel.j.oltmans@noaa.gov; bryan.johnson@noaa.gov

[6] CIRES = Colorado Institute for Research in Environmental Sciences; also at NOAA/Earth Systems Research Laboratory, Boulder, CO 80305; 303-497-3599; owen.r.cooper@noaa.gov

[7] Howard University Department of Physics and Astronomy, 2355 6th St NW, Washington, DC 20059; 202-806-6256; fax- 202-806-5830; ejoseph@howard.edu

[8] Graduate School of Oceanography, 312 CACS Building, University of Rhode Island, Narragansett, RI 02882; 401-874 6715; jmerrill@gso.uri.edu

[9] Department of Physics and Astronomy, 1610 Chapel Dr. East, Valparaiso University, Valparaiso, IN 46383; 219-464-5516; 219-464-5489 (fax); gmorris@valpo.edu

[10] Department of Physics and Astronomy, MS-61, Rice University, 6100 Main Street, Houston, TX 77005; 713-348-5727; 713-348-4150 (fax); shersey@rice.edu

[11] Atmospheric Science Department, University of Alabama in Huntsville, 320 Sparkman Drive, NSSTC 3040, Huntsville, AL 35806; 256-961-7825; 256-961-7751 (fax); mike@nsstc.uah.edu

[12] NASA/Goddard, Wallops Flight Facility, Wallops Island, VA 23337; 757-824-1618; fax 757-824-1036; fjs@osb1.wff.nasa.gov

[13] Environment Canada - Meteorological Services Centre, Downsview, ONT M3H 5T4 416-739-4623; david.tarasick@ec.gc.ca

[14] CNRS, (UMR 5560), Laboratoire d'Aérologie, OMP, 14 Avenue E. Belin, 31400 Toulouse, France; 33-0561- 3327-71 Fax: 33-0561- 332790; thov@aero.obs-mip.fr; camjp@aero.obs-mip.fr

IONS (INTEX Ozonesonde Network Study, 2004): Perspective on Summertime UT/LS (Upper Troposphere/Lower Stratosphere) Ozone over Northeastern North America

24 April 2006

Authors: A M Thompson[1], JBStone[1], JCWitte[2], RBPierce[3], SJ Oltmans[4], ORCooper[5], BFTaubman[1], RBChatfield,[6] BJJohnson[4], EJoseph[7], TLKucsera[2], JTMerrill[8], GAMorris[9], SHersey[10], MJNewchurch[11], FJSchmidlin[12], DWTarasick[13], VThouret[14], JPCammas[14]

ABSTRACT Coordinated ozonesonde launches from IONS (INTEX [Intercontinental Transport Experiment] Ozonesonde Network Study <http://croc.gsfc.nasa.gov/intex/ions.html>) in July-August 2004 provided nearly 300 O_3 profiles from eleven North American sites and the *R/V R H Brown* in the Gulf of Maine. With the IONS period dominated by low-pressure conditions over northeastern North America (NENA), the free troposphere in that region was frequently enriched by stratospheric O_3. Stratospheric O_3 contributions (ST) to the NENA tropospheric O_3 budget are computed through analyses of O_3 laminae (Pierce and Grant [1998]; Teitelbaum et al., 1996), tracers (potential vorticity, water vapor) and trajectories. The ST component ranged from 16 to 34% (mean, 26% $\pm$ 7%) of below-tropopause O_3 over the *R/V R H Brown* and six sites in MI, VA, MD, RI, and Nova Scotia. Analysis of potential vorticity, Wallops ozonesondes (37.9N, 75.5W), and MOZAIC (Measurements of Ozone by Airbus In-service Aircraft) O_3 profiles for NENA airports in JJA (June-July-August) 1996-2004 shows that the stratospheric fraction in 2004 may be typical. Boundary layer (BL) O_3 at Wallops and northeast US sites during IONS also resembled O_3 climatology (JJA 1996-2003). However, statistical classification of Wallops O_3 profiles shows the frequency of profiles with background, non-polluted BL O_3 was greater than normal during IONS.

[1]Penn State Univ, PA; [2]SSAI & NASA/GSFC, MD; [3]NASA/Langley Res Center, VA; [4]NOAA/ESRL, CO; [5] CIRES and NOAA/ESRL, CO; [6] NASA/Ames Research Center, CA; [7]Howard Univ, Wash, DC; [8]Univ of Rhode Island, GSO, RI; [9]Valparaiso Univ, IN; [10] Rice Univ, TX; [11]Univ Alabama-Huntsville, AL; [12]NASA/GSFC/Wallops Flight Facility, VA: [13]Enivronment Canada/MSC, ONT, CA; [14]CNRS-Observatoire du Midi-Pyrenees, FR

1. Introduction.

An ozonesonde network provides consistently placed and timed well-resolved (100-m vertical) observations of tropospheric and stratosphere O_3 that satellite and aircraft platforms do not achieve. Relative ease of operations allows for strategic design and sampling to address the mechanistic requirements of a field campaign. When combined with meteorological fields and P-T-U (pressure-temperature-humidity) radiosonde data, portions of the O_3 budget (schematic in Figure 1) can be deduced without complex models. A good illustration is in the work of Teitelbaum et al. [1994; 1996] where O_3 and water vapor laminae were used to infer gravity and Rossby wave influences in soundings. Similarly, Merrill et al. [1996] used O_3 soundings to identify a wave-breaking mechanism for introducing stratospheric O_3 into the middle and upper troposphere in northern hemisphere summer.

During the ICARTT (International Consortium on Atmospheric Research on Transport and Transformation; Fehsenfeld et al., 2006) and INTEX-NA (Intercontinental Chemical Transport Experiment-North America; Singh et al., 2006) experiments in July-August 2004, coordinated ozonesonde launches were made through IONS (INTEX Ozonesonde Network Study). In this study, we apply the methods of Teitelbaum et al. [1994] and Pierce and Grant [1998] to IONS O_3 profiles to address the following:

- How much tropospheric O_3 in northeastern North America (NENA) originated in the stratosphere?
- Given that July-August 2004 meteorology over NENA was dominated by persistent low-pressure conditions that favor injection of stratospheric O_3 to the troposphere, was stratospheric influence unusually high compared to other years?
- How did 2004 BL (boundary layer) O_3 in NENA compare to climatology?

Experimental and analytical methods are described with a case study in Section 2, followed by two Sections that answer the questions.

2. Observations and Analysis.

2.1 IONS-04. Experiment, Ancillary Data.

The strategic objectives defining IONS were three-fold. (1) With coordinated launches at participating stations (Table 1), measurements were timed for satellite overpasses; sonde data were transmitted within 8 hours of launch to North American and European research aircraft field sites to guide Lagrangian sampling; (2) Geographical coverage of IONS - spanning eight stations and a ship in the Gulf of Maine - was designed for a cross-sectional view from the south central US through maritime Canada. This is illustrated by the ozone mixing ratio "curtain" for 21 July 2004 (Figure 2); (3) Soundings over coastal California, Colorado, Michigan and Ontario allowed study of inter- and cross-continental flows.

Over 290 soundings were taken between 1 July and 15 August 2004, with ECC (electrochemical concentration cell) ozonesondes at all locations. Water vapor is derived from P-T-U profiles measured by radiosondes that fly with the ozonesondes. The ozonesonde-radiosonde system used at Trinidad Head, Houston, Pellston, Huntsville, Narragansett and the *R/V R H Brown* (Thompson et al. [2000]) recorded daily profiles at 1-s frequency, generally during overpasses of the Aqua and Aura satellites. The latter was launched on 15 July 2004 with four O_3 instruments (<http://aura.gsfc.nasa.gov>). Other IONS sites launched sondes 1-3 times/week. Images of all IONS O_3 profiles are at: <http://croc.gsfc.nasa.gov/intex/ions.html>.

For comparisons with O_3 climatology over NENA, MOZAIC profiles (Measurements of Ozone and Water Vapour by Airbus In-service Aircraft; <http://aeropc35.aero.obs-mip.fr>) and surface O_3 from an EPA network, <http://epa.gov/aqspub1/site.html>, are used. Satellite data, trajectory-enhanced meteorological fields and imagery as well as standard analyses (e.g. potential vorticity, potential temperature; all determined with GEOS-4, <http://croc.gsfc.nasa.gov/intex>) are used to interpret O_3 and P-T-U profiles.

All trajectories are run with a kinematic version of the Goddard trajectory model [Schoeberl and Newman, 1995] using GEOS-4 analyses.

2.2 Evaluation of Boundary-Layer and Stratospherically-influenced Ozone

Two terms in the tropospheric O_3 budget are foci for the present study: boundary-layer (BL) and stratospherically-influenced (ST) ozone. Except for a few low-level inversions (200-300 m) and several boundary layers that extended above 1 km, the P-T-U profiles show that the BL over IONS stations was between 0.5 to 1 km. Lower and upper limits for computed BL O_3 are based on integration from the surface to 0.5 and 1 km, respectively, with column amounts given in DU (Dobson Units; 1 DU = 2.69×10^{16} cm^{-2}). For comparison to O_3 measured at surface monitoring locations, IONS "surface O_3" is determined by averaging O_3 measured by the sonde from the surface to 100 m. Including more than the lowest 2-3 values allows for the sonde response time and occasional interferences with ECC O_3 readings at the surface. A similar calculation gives surface O_3 from the MOZAIC profiles.

ST O_3 is evaluated through a combination of tracers and an analytic method designated here as lamina-labeling or the PT method, after Pierce and Grant [1998] and Teitelbaum and coworkers [1994; 1996]. The PT method is based on the observation made by Holton [1987] and others [Dobson, 1973; Reid and Vaughan, 1991], that most soundings show a relatively constant O_3 profile within the troposphere or stratosphere except where regional perturbations lead to stable layers that deviate significantly from the mean. Newell et al. [1999] made the same observation with ozone from aircraft profiling. In Figure 2, a typical set of IONS soundings, all the profiles illustrated display a relatively constant mixing ratio between the BL and the upper troposphere/lower stratosphere (UT/LS). Note that the terms "UT" and "UT/LS" are both used to describe the region below the tropopause, where the latter is determined by conventional thermal or ozone-based definitions. Huntsville and Beltsville display a distinct tropopause; the region below the sharp red-to-brown transition can be considered as UT. The transitional character of a 2-km thick region shaded red-orange-yellow makes "UT/LS" a more appropriate designation for the other stations. For Sable Island, the mean tropospheric mixing ratio is 40-45 ppbv, corresponding to light blue-light green in Figure 2. For all profiles except Sable Island, the mid-tropospheric mean O_3 is ~75 ppbv, green-to-yellow shading, and the UT/LS mean is ~100 ppbv O_3, orange in Figure 2.

<u>2.2.1 Standard PT Method: Rossby-Wave Identification from Lamina-labeling</u>

Lamina-labeling refers to attribution of the layers illustrated in Figure 3 to two processes: Rossby waves (RW) and gravity waves (GW), as described by Teitelbaum et al. [1994] and Pierce and Grant [1998]. For a given set of soundings, normalized O_3 gradients are compared to gradients in the normalized potential temperature (θ), calculated from the P-T-U data. The normalized θ gradients appear as dotted lines in Figure 3. A high degree of correlation between the O_3 and θ, signified by dashed lines extending to > 0.7, is attributed to GW [Pierce and Grant, 1998; Teitelbaum et al., 1994]. Regions where the O_3-θ correlation is low, $\leq$ 0.3 absolute, are identified with RW.

In the NENA region, which was subject to recurring low-pressure systems during IONS, tropospheric laminae due to RW are identified with filaments from wave-breaking, ie stratospheric influence, provided there is supporting evidence from trajectories and/or tracers, H_2O or pv (compare the GW studies of Teitelbaum et al., 1994). In the standard lamina-labeling method of Pierce and Grant [1998], RW and GW-influenced layers are determined from each ozonesonde-radiosonde set as follows:

<u>Step 1:</u> Identify the tropopause, both TTP (thermal tropopause) and OTP, an ozone-defined tropopause. The latter is determined with the method of Browell and coworkers [Browell et al., 1996a,b; Fenn et al., 1999] where the OTP is the first 100 ppbv O_3 mixing ratio point below a specified rate of stratospheric ozone decline. In Figure 2 this generally corresponds to the

first occurrence of 100 ppbv ozone below the brown color.

Step 2: Locate laminae in O_3 through normalization to a running mean and potential temperature laminae, θ, from the radiosonde data. For Sable Island, pronounced laminae at 13 km, 10.5 km and 4 km appear as solid lines and positive deviations from the 40-45 ppbv O_3 mean in Figure 3a. The θ deviations are dotted lines. Negative deviations define O_3 laminae at 13.5 km, 12.2 km, 9 km and 2.5 km. Positive O_3 deviations for the *R/V R H Brown* and Narragansett (Figure 3b) are significant at 9 km, more red than orange, and at 3-4 km, more yellow-orange than the 75 ppbv O_3 mean.

Step 3: Run PT analysis for GW and lower and upper RW limits. Correlations are given by dashed lines in Figure 3. The "low-correlation" criterion of Pierce and Grant [1998] is raised to $\pm$0.4 and lowered to $\pm$0.2 to bracket, respectively, maximum and minimum RW.

Step 4: Compute ozone in each RW layer (RW in Dobson Units)

Step 5: Add all such layers within profile to obtain total RW, absolute and fraction, within the O_3 profile from surface to OTP or TTP.

Two sources of uncertainty affect the RW determination in Steps 1-5. First, not all O_3 profiles display sufficiently stable layers to give meaningful results with lamina-labeling. This eliminates profiles affected by active mixing, presumably from both surface and the stratosphere. Fewer than 10% of the NENA IONS profiles are excluded by this criterion. Second, O_3 and P-T-U data between 750 and 200 hPa during IONS were such that the O_3-θ gradient correlations often shifted near the edges of the RW range, e.g. the blue-dashed correlation curve for *R/V R H Brown* near 9 km in Figure 3b. This uncertainty is estimated as equivalent to a typical O_3 column depth in 0.5 km ($\pm$ 2-3 DU).

Figure 3 displays OTP and TTP and illustrates how lamina-labeling applies to three IONS profiles for 21 July 2004. The maximum and minimum RW ranges are shaded green in Figure 3. Table 2 summarizes the BL and RW amounts and fractions for six NENA stations on 21 July, along with analysis of the same day's soundings over Pellston, Michigan. Three RW values are given, two based on the minimum and maximum RW criteria for an O_3-defined tropopause (OTP) and one derived from the standard RW criterion, 0.3 absolute for the low-correlation limit [Teitelbaum et al., 1994; Pierce and Grant, 1998], with a thermally defined tropopause (TTP).

Table 2. Ozone on 21 July 2004 at IONS locations, with stratospherically influenced tropospheric O_3 (ST) and BL O_3. Pells = Pellston; Belts = Beltsville; WFF = Wallops Island; Narra= Narragansett; *RHB* = *R/V R H Brown*; Yarm = Yarmouth

21 July 2004	Pells	Belts	WFF	Narra	*RHB*	Yarm	Sable Is
O_3 (to OTP, DU)	42	67.3	69.9	37.3	43.6	52.3	29.7
OTP (km)	13.9	13.8	12.4	7.0	9.5	12.7	14.1
RW-OTP (minimum,* DU)	5.8	6.6	14.2	0.0	0.0	14.6	2.7
RW-OTP (maximum,* DU)	15.7	32.2	22.1	6.7	12.6	21.2	12.8
RW-OTP (minimum fraction)	0.14	0.10	0.20	0.0	0.0	0.28	0.09
RW-OTP (max. fraction)	0.37	0.48	0.32	0.18	0.29	0.41	0.43
O_3 (to TTP, DU)	42.0	49.4	77.2	85.0	60.0	52.3	28.2
TTP (km)	13.4	10.9	14.2	13.9	12.7	12.8	13.9
RW-TTP (fraction)	0.23	0.15	0.30	0.33	0.16	0.34	0.16
BL (surface to 0.5 km, DU)	1.5	3.4	2.5	2.7	1.6	2.5	1.6
BL (0.5 km fraction to OTP)	0.04	0.05	0.05	0.07	0.04	0.05	0.05

For 21 July 2004, the TTP for the NENA sites is 11-14 km. For all but two stations (*R/V R H Brown* and Narragansett), the OTP and TTP difference is less than 3 km. The greatest divergence between the OTP and TTP, nearly 7 km at Narragansett, leads to a large difference between tropospheric O_3 column and RW fractions computed with the two criteria. The tropospheric O_3 is 37 DU integrated to the OTP (at 7.0 km) whereas total O_3 is 85 DU to 13.9 km. This is equivalent to an RW minimum value of 0 (maximum of 0.18), based on the OTP. The conventional RW criterion, correlation $\leq |0.3|$ with the TTP, gives a value for RW O_3 = 0.33 at Narragansett.

2.2.2 Modified PT Method: ST Ozone from PT-Identified Rossby-Waves

The final step in lamina-labeling as applied to IONS data is the use of tracers to assign the amount of O_3 identified as RW to stratospheric (ST) ozone within the profile.

This has been done in two ways. A combination of H_2O within the profile, potential vorticity (pv) and back-trajectories is used to scale RW O_3 amounts within each profile. In the 21 July example, RW is equivalent to ST at Narragansett and *R/V R H Brown* because the RW-labeled laminae correspond to relatively dry segments of the profile. This is indicated by the degree of under-saturation in Figure 4, where dew-point temperature, DPT, is much less than the temperature, T. For Sable Island the DPT-T comparison suggests near-saturation, not dry stratospheric air, throughout the profile. Accordingly, for 21 July, the RW to ST scaling is 1.0 for Narragansett and *R/V R H Brown* and 0.0 for Sable Island.

This interpretation is reinforced by pv (in the analyses here, Ertel's pv, computed from GEOS-4) and the origins of back-trajectories. The following summarizes two analyses that point to stratospheric origins at Narragansett and the *R/V R H Brown*:

1) Back trajectories initiated from the mid-troposphere over the *R/V R H Brown* and Narragansett show O_3 in those air parcels originating over Canada at 60^0N (Figure 5) where the trajectory-mapped pv corresponds to stratospheric values, > 3 pvu. The levels for the O_3 soundings at which the air parcel trajectories were initialized correspond to altitudes of lamina associated with RW. The coherence of the trajectories and pv for Narragansett and the *R/V R H Brown* supports identification of the corresponding segments of the profile with ST O_3 (dashed blue and black lines within the right-hand shaded band at 6-9 km in Figure 3b) on 21 July 2004.

2) The trajectory-mapped pv in Figure 5 displays a filament with elevated pv (> 3 pvu at ~11 km in Figure 5) over the *R/V R H Brown* and Narragansett, that extends south to the mid-Atlantic region. Meteorological fields, e.g. P-T-U fields for 21 July 2004 at <http://croc.gsfc.nasa.gov/intex>, show this narrow filament to be cooler and drier than the surrounding regions. To the east of the filament is a transition toward subtropical pv levels (denoted purple in Figure 5) that includes Yarmouth (red spot in western Nova

Scotia; cf Table 1) and Sable Island (easternmost red spot in Figure 5). As the transition suggests, Yarmouth's 10-km O_3 mixing ratio (~ 80 ppbv; Figure 2) is less than the 10-km O_3 mixing ratio at Narragansett and the *R/V R H Brown*, both > 100 ppbv. Lamina-labeling is sufficiently sensitive to pick up RW character in the Yarmouth sounding (Table 2) and the suppressed DPT confirms stratospheric influence below 12 km.

Although a matrix of H_2O, pv and back-trajectories usually gives a coherent basis for evaluating RW in each profile, the information is sometimes contradictory. Compared to distances among IONS stations and to vertical resolution in laminae, the meteorological fields used for pv and trajectories are relatively coarse. For example, plots of UT/LS pv vs O_3 mixing ratio (evaluated at 150 hPa, Figure 6a) show pv and O_3 tracking one another but the correlation is inconsistent (Figure 6b). Water vapor turns out to have the most consistent relationship with O_3 and a second method for converting RW to ST O_3 was developed by using the O_3-H_2O anti-correlation for each site. The scale factor is based on the fraction of RW laminae in the entire dataset that are sufficiently dry for a stratospheric signature to be credible. Sable Island has the lowest scale factor, 0.5; for the other sites, the conversion of RW O_3 to ST O_3 is ~0.7-0.9 (Table 3).

3. Results: 2004 IONS ST and BL Ozone

Section 3.1 summarizes statistics for BL and ST O_3 over all the NENA IONS soundings. These are discussed with reference to ancillary observations during the IONS period in Section 3.2.

3.1 Composite ST and BL Ozone Statistics during IONS

The Narragansett OTP-TTP discrepancy described in Section 2.2.1 is not typical for IONS. Over the entire set of profiles analyzed by lamina-labeling, the mean OTP-TTP difference is 0.3 km. Thus, for statistical analysis of ST and BL fractions over IONS profiles as a whole, a single definition (OTP) is used. Averaged parameters for OTP, O_3 column amounts, ST and BL fractions appear in Table 3.

Table 3. Maximum and minimum ST and BL O_3 amounts and fractions for IONS-04 soundings (1 July -13 August 2004).

IONS-04	Pells	Belts	WFF	Narr	*RHB*	Yarm	Sable	Trini
O_3 (to OTP, DU)	39.9	47.1	47.5	44.7	45.6	45.4	40.7	40
OTP (km)	11.7	12.1	13.8	12.2	12.5	12.6	14.8	12.6
RW-OTP (minimum,* DU)	9.8	14.3	13.6	12.1	8.4	12.5	8.7	9.8
RW-OTP (maximum,* DU)	17.4	21.8	25.2	20.4	19.3	20.9	17.7	20.3
RW-OTP (minimum fraction)	0.24	0.30	0.29	0.27	0.18	0.28	0.21	0.24
RW-OTP (maximum fraction)	0.44	0.46	0.53	0.46	0.42	0.46	0.43	0.51
RW to ST Scale Factor	0.89	0.75	0.82	0.72	0.85	0.67	0.50	0.70
ST O_3 (DU) **	12.1	13.5	16.0	11.7	11.8	11.1	6.6	10.5
ST O_3 (fraction)	0.30	0.28	0.34	0.26	0.26	0.25	0.16	0.26
O_3 (to 15 km, DU)	59.6	58.2	49.6	57.2	60.6	52.8	47.5	49.7
BL (surface to 0.5 km, DU)	0.87	2.6	2.0	1.8	1.9	1.7	1.2	1.0
BL (0.5 km fraction to OTP)	0.02	0.06	0.04	0.04	0.04	0.04	0.03	0.025
BL (surface to 1.0 km, DU)	2.6	5.0	4.4	4.4	3.9	3.8	2.6	2.4
BL (1.0 km fraction to OTP)	0.07	0.11	0.09	0.10	0.09	0.08	0.06	0.06
Surface O_3 (0-100 m, ppbv)	34	66	47	46	30	35	25	18

* Minimum, maximum determinations defined in Section 2.2 and Figure 3.

** Average from scaling RW-OTP maximum and minimum to ST O_3

Amounts and fractions of ST O_3 are similar among NENA sites, except for Sable, where the absolute amount is about half that for the other locations and the fraction is less by 0.10 absolute. The mean ST fraction for the other NENA sites is 0.28 ($\pm$0.03). At Trinidad Head and Pellston, two non-NENA sites with daily soundings, ST O_3 amounts, both absolute and fractional, are similar to the NENA budgets.

The BL O_3 amounts (0 - 0.5 km integrals) for Trinidad Head and Pellston, sites usually remote from pollution, are roughly half the amount at NENA locations. Sable Island is also low in BL O_3 because it usually had marine, southeasterly near-surface flows. The most urban site, Beltsville, has the highest BL O_3, 2.6 DU to 0.5 km, ~25% greater than the other locations listed in Table 3.

3.2 Observations Related to IONS ST and BL Ozone.

Three additional analyses put 2004 ST and BL O_3 into perspective.

[A] Ozone at 10-15 km. To further minimize ambiguity in the interpretation of ST O_3

due to tropopause definitions, the integrated 10-15 km O_3 column is computed (average value in Table 3). This can be considered a geometrically prescribed UT/LS O_3 amount. Figure 7a shows that the fraction of O_3 in the 10-15 km layer, relative to surface-to-15 km O_3, compares well with the below-tropopause RW fraction. RW O_3 fraction averages 0.35 among the six NENA sites; with scaling by H_2O, the ST O_3 fraction is 0.27, except for Wallops (ST O_3= 33%) and Sable Island, where 50% down-scaling denotes less stratospheric influence.

[B] MOZAIC LTO (landing and takeoff) profiles [Thouret et al., 1998]. These are normally measured below 200 hPa (~ 11 km) and are useful for characterizing UT/LS O_3 when limited sonde data are available (Cooper et al., 2005, 2006; Section 4). The 9-11 km O_3 segments from MOZAIC profiles during JJA 2004, mostly from LTO at New York and Washington, are compared with IONS segments between 9 and 11 km at NENA locations (Figure 7b). Strong similarity in the fractions suggests that characteristics of IONS sonde O_3 profiles, e.g. fractions of ST O_3, apply to the larger NENA region.

[C] Potential Vorticity. The mean tropopause height (OTP in Table 3) for the six NENA sites corresponds to 150 hPa. A sequence of O_3 averaged between 10 and 15 km (mid-point ~ 150 hPa) from two IONS sites shows typical tracking with pv (Figure 6a). Stratospheric O_3 is identified with O_3 > 150 ppbv and pv > 3 pvu. When mean O_3 is ~100 ppbv and pv is less than 2 pvu, the PT method may show RW segments and ST O_3. The positive correlation between pv and O_3 at the sites in Figure 6b is typical of IONS data as are recurring episodes of stratospheric influence throughout July-early August 2004. Thus, pv is used as a tracer for stratospheric influence when profile data are not available above 200 hPa (Section 4). Figure 6a may suggest a tendency toward lower pv (and lower O_3 mixing ratio) as summer progresses. This is consistent with trajectory analysis of MOZAIC LTO profiles in which Cooper et al. [2006] conclude that stratosphere-to-troposphere injections occur throughout North American summer but with declining O_3 flux over time.

Figure 8 presents O_3 surface mixing ratio for six NENA sites during IONS and for four MOZAIC airports (surface to 100 m means in JJA 2004). Because profile data represent 20 seconds of sampling, surface readings from O_3 monitoring sites (continuous readings) near each city are also presented. The latter are daytime averages from sites reporting to the EPA (<www.epa.gov/aqspub1/site.html>]. The marine locations, *R/V R H Brown*, Sable Island and Yarmouth on the western Nova Scotia coast, are in the 25-35 ppbv range. This resembles O_3 in the lowest layer shown for 21 July 2004 in Figure 2. On average for the IONS period, Wallops and Narragansett surface O_3, at 47 and 46 ppbv (Figure 8), respectively, are more affected by pollution than Yarmouth and the *R/V R H Brown*. The near-urban location, Beltsville, averaged 66 ppbv for the IONS period, greater than the nearby MOZAIC (Washington-Dulles) and Baltimore-Washington EPA data (JJA mean O_3 = 45 ppbv; Figure 8). On average, O_3 mixing ratios at IONS sites are lower than MOZAIC surface measurements. Besides the latter being more urban, aircraft profiling usually occurs a few hours after the sondes, at mid-afternoon to early evening, when surface O_3 is near its daily maximum.

4. 2004 Ozone and Climatology.

To compare ozone from IONS with recent climatology (1996-2004), Wallops sondes and MOZAIC profiles are used to examine the free troposphere and UT/LS (Section 4.1) and BL and surface ozone (Section 4.2).

4.1 UT/LS Ozone and Potential Vorticity over NENA (1996-2004)

Wallops is the only NENA IONS station with a multi-year sounding record (Newchurch et al., 2003). Normal sampling frequency is weekly, usually mid-week within a couple hours of local noon. ST and BL O_3, as in Table 3, are computed from Wallops soundings for JJA 1996-2004. Figure 9a, the time-series of mean ST O_3, shows that 2004 is not unusual in the recent climatology. Nor is the fraction of UT/LS ozone in the 10-15 km layer, also displayed in Figure 9a. Wallops, at the southern end of the region designated as NENA in this study, may not be representative of the northeastern US or maritimes. MOZAIC LTO O_3 profiles are used as a reference for the mid-upper troposphere (300-200 hPa, 9-11 km). A time-series of 1996-2004 MOZAIC and Wallops 9-11 km O_3 column amounts (Figure 9b) shows remarkable consistency within each data set, though MOZAIC averages 2-3 DU greater in absolute O_3 amount and 30-40% fractionally. For 2004, the standard deviation of both data sets is small and the 9-11 km column and fractional amounts are identical, as implied by Figure 7b.

To compensate for limited pre-2004 O_3 data above 200 hPa over NENA, statistics for pv at 150 hPa (cf Thompson et al., 1999) are used to determine whether ST O_3 fractions during IONS might be unusual. Figure 10 displays probability distributions of daily epv for JJA in 1996-2004 over the locations of Pellston and the six NENA sites analyzed in Tables 2, 3 and Figures 7 and 8. The 2004 distribution (red in Figure 10) is somewhat broader than other years, with more values > 10 pvu. However, standard statistical tests do not show 2004 to be anomalous. Thus, pv statistics at IONS sites, like the Wallops O_3 analysis, suggest that 2004 was a typical year for UT/LS O_3 over NENA.

4.2 BL Ozone at NENA IONS Sites in the 1996-2004 Context

The synoptic conditions of NENA during IONS appeared to be anomalous when compared to climatology. Parameters like temperature, cloud cover, precipitation amount (supplementary data based on analyses provided by <http://www.cdc.noaa.gov>) describe a summer that was cooler and wetter than normal. Ozone at the surface, based on daily maximum O_3 values from continuous data over JJA 2004 at EPA sites around Washington, DC; Baltimore and Philadelphia, were in the lowest 25-30% of values recorded during the period 1984-2004 (not shown).

Figure 11 shows the surface (lowest 100 meter averages) O_3 mixing ratio for

Wallops and MOZAIC NENA locations, averaged over JJA from 1996 to 2004. EPA daytime mean O_3 averaged over the 5 locations of Figure 8 is also displayed. The EPA O_3 resembles the Wallops pattern, with the two means within 7 ppbv of one another, Wallops higher in all but two years. Wallops surface O_3 during 2004 does not stand out in the record, but mean surface O_3 is 15% lower than the 2001 and 2002 Wallops means. The O_3 column (to 2.0 km) at Wallops was 8.7 DU in 2004 compared to 10.5+/-0.5 DU for 1996-2003. The MOZAIC surface O_3 data are not always consistent with Wallops and EPA records. It isn't clear why MOZAIC O_3 is so low in 1998 and 2000.

Means are not always the most effective way to evaluate O_3 profiles, as Diab et al. [2003, 2004] demonstrated with statistical classification of sonde and MOZAIC data over the Johannesburg (25S, 28E) area. Like the IONS sites, Johannesburg is a region of mixed pollution, marine and stratospheric influences. Prototype O_3 profiles for the middle and lower troposphere over Johannesburg, corresponding to distinct meteorological situations, emerged from agglomerative hierarchical clustering [Diab et al., 2004] of the data. Following the latter study, we classified the O_3 mixing ratio measurements taken from Wallops sondes up to 200 hPa for JJA in 1996-2004 (148 total sondes in Figure 12). Three prototypes are distinguished in clustered distributions of the Wallops profiles. Category 1, resembling marine tropical air, is minimally polluted throughout the profile. A second category is relatively unpolluted in the lowest 5 km but displays higher O_3 mixing ratio in the middle troposphere. Category 3 is distinguished with high pollution frequency below 5 km. The BL O_3 means are: 44 ppbv, Category 1; 46 ppbv, Category 2; 56 ppbv, Category 3. The yearly distribution for Wallops, displayed as fractions of each year's JJA profile count, appears in Figure 12. Wallops in 2004 was unique in having 20% of its profiles in Category 1, compared to five years in 1996-2004 with 5-10% occurrence and three years with none. Only 30% of Wallops profiles (a minimum value, met only in one other year) were in Category 3 (high-pollution) for 2004, compared to > 40% of JJA profiles in five summers in the 1996-2003 period.

5. Discussion and Summary

5.1 Comparison with Other Ozone Studies

Prior to INTEX and ICARTT, the importance of stratospheric O_3 on the mid-latitude free troposphere was demonstrated with aircraft data (Parrish et al., 2000; Esler et al., 2003) or by trajectories related to surface O_3 [Moody et al., 1995; Merrill and Moody, 1996]. These observations show more stratospheric flux during springtime (as expected from models, e.g. Meloen et al., 2003; Olsen et al., 2004) but the frequency of stratospheric encounters at other seasons has also been remarkable. Aircraft O_3 profile classifications [Browell et al., 1996a] displayed stratospheric influences in 40-50% (frequency, not mass content) of the mid-latitude O_3 profiles above 300 hPa during late summer and early fall. In NENA flights designed to sample aircraft and surface-influenced pollution in early fall 1997 [Singh et al., 1999; Thompson et al., 1999], stratospheric O_3 was encountered on 40% of flights. Cho et al. [1999] detailed gravity wave influences on the UT/LS and showed that pollution transport can be modified by stratospheric capping [Cho et al., 2001]. Stratospheric influences during INTEX and ICARTT were observed far downwind of NENA, in O_3 and CO sampling at the Pico (Azores) location [M. Val Martin et al., Personal communication, 2006], where pollution impacts from NENA export were more prominent.

The IONS network is the first time that NENA sondes have been distributed with sufficient temporal and spatial coverage to permit evaluation of day-to-day O_3 budgets. The data have been used to evaluate regional and global models [Al-Saadi et al., 2005; Pfister et al., 2005b; Tarasick et al., 2005] and to assess O_3 contributions with the FLEX-PART trajectory approach [Cooper et al., 2006]. In the latter study, combined MOZAIC and IONS profiles for some sites make direct comparison between the ST O_3 analyses from the PT (lamina-labeling) method and the FLEXPART budgets impractical. However, for

NENA sites where comparisons are straightforward (Sable, Narragansett and the *R/V R H Brown*), FLEXPART assigns a 10-15% fraction of below-tropopause O_3 to stratospheric influence. Like lamina-labeling, FLEXPART shows Sable Island to be less influenced by stratospheric O_3 than the other two locations. The FLEXPART budget is based on an effective tropopause height that averages 1-2 km lower than OTP in Table 3. This would be equivalent to 3-4 DU less ST O_3 (Table 3) or 10% less absolute ST O_3, bringing the lamina-labeling and FLEXPART estimates into agreement with one another.

5.2 Summary

Using several hundred O_3 soundings collected through IONS during the INTEX-NA and ICARTT experiments in July-August 2004, the present study evaluates two terms in the budget, BL and ST O_3 (Figure 1). The emphasis is on ST O_3 determined with lamina-labeling and tracers. Over five NENA sites (Wallops, Beltsville, Narragansett, Yarmouth and the *R/V R H Brown*) the mean ST O_3 fraction is 26 ($\pm$ 7%), a value that also holds for Pellston ~ 1000 km west of New England. Sable Island, east of the region affected by low-pressure in July and August 2004, has lower ST O_3, 16% over the IONS period. These amounts are equivalent to stratospherically influenced tropospheric O_3 determined from FLEXPART [Cooper et al., 2006] when tropopause differences between the two methods are taken into account.

BL column O_3, to 0.5 km, and surface O_3 were also determined from the 2004 IONS soundings. A climatological perspective on IONS is given by Wallops sondes, EPA data from the NE US and NENA MOZAIC landing/takeoff O_3 profiles from 1996-2004. The BL in 2004 over Wallops showed a very low frequency of O_3 surface pollution for JJA in the 1996-2004 period. However, on average, Wallops surface O_3 in 2004 was close to the 9-year average. The MOZAIC and EPA surface O_3 records for 2004 were not unusual.

For Wallops, the O_3 profiles and pv tracer showed that stratospheric influences in 2004 were typical. Potential vorticity statistics for IONS locations during JJA 1996-2003 suggest that a 15-30% ST O_3 value over NENA in summer could be a general result. Although episodes of mid-latitude ST O_3 in summer [Moody et al., 1995; Merrill et al., 1996] or fall [Cho et al., 1999; Thompson et al., 1999] have been reported over eastern North America and the North Atlantic, there has not been enough profile data to evaluate the O_3 budget. The statistics presented here show that summertime stratospheric influences in the North American troposphere may be more prevalent than previously considered. This conclusion is based on the unique temporal and spatial coverage afforded by the IONS network, MOZAIC and the long-term sounding record at Wallops.

Acknowledgments. We are grateful to Chief Scientist T. S. Bates, J. E. Johnson, D. Hamilton (NOAA/PMEL) and the crew of the *R/V Ronald H. Brown*. Students J. F. Liesch, V. Davis, M. Taylor, O. Hylton, L. M. Zamora and the NASA/Goddard Directors Discretionary Fund provided the Beltsville soundings. Thanks to J. L. Moody and A. Klepac (U. Virginia) for the Pellston sondes. Useful comments were made by R. R. Dickerson (U. Maryland), K. E. Pickering (NASA/Goddard), J. L. Moody. NASA's Tropospheric Chemistry Program, NOAA's CMDL and Aeronomy Labs and Environment Canada were the primary IONS sponsors. Analysis by AMT and JBS, whose MS Thesis is based on this work, is supported by NASA Aura Validation and Penn State's Meteorology Department. Thanks to C. A. Piety (U. Maryland) and M. P. McEvoy (Penn State) for EPA data and to S. K. Miller (Penn State) for graphics assistance. All IONS data and images reside at the ICARTT/INTEX archive: <http://cloud1.arc.nasa.gov>.

Table 1. IONS Stations in 2004.

R/V R H Brown Operating from Portsmouth, NH	
Beltsville, MD, USA	39.04 N, 76.52 W
Boulder, CO, USA	40.30 N, 105.20 W
Egbert, Ont, CAN	44.23 N, 79.78 W
Houston, TX, USA	29.72 N, 95.40 W
Huntsville, AL, USA	35.28 N, 86.58 W
Narragansett, RI, USA	41.49 N, 71.42 W
Pellston, MI, USA	45.57 N, 84.68 W
Sable Is., N.S., CAN	43.93 N, 60.01 W
Trinidad Head, CA, USA	40.80 N, 124.15 W
Wallops Is, VA, USA	37.85 N, 75.50 W
Yarmouth, N.S., CAN	43.87 N, 66.12 W

REFERENCES.

Al-Saadi, J., R. B. Pierce, T. D. Fairlie, C. Kittaka, T. K. Schaack, T. Zapotocny, D. R. Johnson, M. A. Avery, A. Thompson, R. C. Cohen, J. E. Dibb, J. H. Crawford, D. F. Rault, J. J. Szykman, R. Martin, Lagrangian characterization of the sources and chemical transformation of airmasses observed by the NASA DC-8 during ICARTT/INTEX-A, *Eos Trans. AGU*, **86**(52), Fall Meet. Suppl., Abstract A53D-04, 2005.

Bloom, S., A. da Silva, D. Dee, M. Bosilovich, J.-D. Chern, S. Pawson, S. Schubert, M. Sienkiewicz, I. Stajner, W.-W. Tan, M.-L. Wu, Documentation and Validation of the Goddard Earth Observing System (GEOS) Data Assimilation System - Version 4 . Technical Report Series on Global Modeling and Data Assimilation 104606, 2005.

Browell, E. V., et al., Large-scale air mass characteristics observed over the Western Pacific during summertime, *J. Geophys. Res.*, 101, 1691-1712, 1996a

Browell, E. V., et al., Ozone and aerosol distributions and air mass characteristics over the South Atlantic Basin during the burning season, *J. Geophys. Res.,* **101**, 24043-24068, 1996b.

Cho, J. Y. N., et al,, Observations of convective and dynamical instabilities in tropopause folds and their contribution to stratosphere-troposphere exchange, *J. Geophys. Res.*, **104**, 21549-21568, 1999.

Cho, J. Y. N., R. E. Newell, E. V. Browell, W. B. Grant, C. F. Butler, M. A. Fenn, Observation of pollution plume capping by a tropopause fold, *Geophys. Res. Lett.*, **28**, 3243-3246, 2001.

Cooper, O. R., et al., A springtime comparison of tropospheric ozone and transport pathways on the east and west coasts of the United States, *J. Geophys. Res.,* **110,** D05990, doi: 10.129/2004JD005183, 2005.

Cooper, O. R., et al., Large upper tropospheric ozone enhancements above mid-latitude North America during summer: In situ evidence from the IONS and MOZAIC ozone monitoring network, *J. Geophys. Res.,* submitted, 2006. Also *AGU, Eos Trans.*, **86** (52), Fall Meeting Suppl. Abs 53-05, 2005.

Diab, R. D., A. Raghunandran, A. M. Thompson, V. Thouret, Classification of tropospheric ozone profiles over Johannesburg based on MOZAIC aircraft data, *Atmos. Chem. Phys.*, **3**, 713-723, 2003.

Diab, R. D., A. M. Thompson, K. Mari, L. Ramsay, G. J. R. Coetzee, Tropospheric ozone climatology over Irene, South Africa from 1990-1994 and 1998-2002, *J, Geophys. Res.,* **109,** D20, D20301, doi: 10.129/ 2004JD004293, 2004.

Dobson, G. M. B., The laminated structure of the ozone in the stratosphere, *Q. J. Royal. Meteor. Soc.,* **99,** 599-607, 1973.

Esler, J. G., P. Haynes, K. S. Law, H. Barjat, K. Dewey, J. Kent, S. Schmitgen, N. Brough, Transport and mixing between airmasses in cold frontal regions during Dynamics and Chemistry of Frontal zone (DCFZ), *J. Geophys. Res.,* **108,** (D4) 4142, doi: 10.129/2001JD001494, 2003.

Fehsenfeld, F. C., et al., International Consortium for Atmospheric Research on Transport and

Transformation (ICARTT): North America to Europe: Overview of the 2004 summer field study, *J. Geophys. Res.,* to be submitted, 2006.

Fenn, M.et al., Ozone and aerosol distributions and air mass characteristics over the south Pacific during the burning season, *J. Geophys. Res.,* **104,** 16,197-16,212, 1999.

Holton, J. R., The production of temporal variability in trace constituent concentrations, in *Transport processes in the middle atmosphere,*, G. Visconti and R. Garcia, eds., D. Reidel Pub., Dordrecht, pp 313-326, 1987.

Meloen, J., et al., Stratosphere-troposphere exchange: A model and method intercomparison, *J. Geophys. Res.,* **108**, D12, 8526, doi: 10.129/2001JD001494, 2003.

Merrill, J. T., J. L. Moody, S. J. Oltmans, H. Levy II, Meteorological analysis of tropospheric ozone profiles at Bermuda, *J. Geophys. Res.,* **101**, 29201-29212, 1996.

Moody, J. L., S. J. Oltmans, H. Levy II, J. T. Merrill, Transport climatology of tropospheric ozone: Bermuda, 1988-1992, *J. Geophys. Res.,* **100**, 7179-7194, 1995.

Morris, G. A., et al., Alaskan and Canadian forest fires exacerbate ozone pollution in Houston, Texas, on 19 and 20 July 2004, *J. Geophys. Res.,* doi: 10.129/2006JD007090, in press, 2006.

Newchurch M. J., M. A. Ayoub, S. Oltmans, B. Johnson, F. J. Schmidlin, Vertical distribution of ozone at four sites in the United States, *J. Geophys. Res*., **108** (D1), 4031, doi:10.1029/2002JD002059, 2003.

Newell, R. N., V. Thouret, J. Y. N. Cho, P. Stoller, A. Marenco, H. G. Smit, Ubiquity of quasi-horizontal layers in the troposphere, *Nature,* **398,** 316-319, 1999.

Olsen, M. A., M. R. Schoeberl, A. R. Douglass, Stratospheric-tropospheric exchange of mass and ozone, *J. Geophys. Res.,* **109,** D24114, doi: 10.129/2004JD005186, 2004.

Pfister G., P. G. Hess, L. K. Emmons, J.-F. Lamarque, C. Wiedinmyer, D. P. Edwards, G. Pétron, J. C. Gille, G. W. Sachse, Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data, *Geophys. Res. Lett.*, 32, L11809, doi: 10.1029/2005GL022995, 2005a.

Pfister G., L. K. Emmons, P. G. Hess, J.-F. Lamarque, D.P. Edwards, A.M. Thompson, D.J. Wuebbles, R.L. Herman, C.R. Owen, R. Honrath, M. Val Martin, G.W. Sachse, M. Avery, J.T. Randerson, Implications of North American boreal fires on air quality and composition in nearby and remote regions, *Eos Trans. AGU*, **86**(52), Fall Meet. Suppl., Abstract A21B-0847, Fall AGU, 2005b.

Pierce, R. B., and W. B. Grant, Seasonal evolution of Rossby and gravity wave induced laminae in ozonesonde data obtained from Wallops Island, Virginia, *Geophys. Res. Lett.,* **25,** 1859-1862, 1998.

Reid, S. J., and G. Vaughan, Lamination in ozone profiles in the lower stratosphere. *Q. J. R. Meteorol. Soc.,* 117, 1991.

Schoeberl, M. R., and P. A. Newman, A multiple-level trajectory analysis of vortex filaments, *J. Geophys. Res.,* **100**, 25801-25815, 1995.

Singh, H. B., A. M. Thompson, H. Schlager, The SONEX and POLINAT Experiments: An overview, *Geophys. Res. Lett.*, **27,** 3255-3258, 2000.

Singh, H. B., W. H. Brune, J. H. Crawford, D. J. Jacob, Overview of the summer 2004 Intercontinental Chemical Transport Experiment - North America (INTEX-NA), *J. Geophys. Res.,* to be submitted, 2006.

Tarasick, D. W., et al., Comparison of Canadian air quality forecast models with tropospheric ozone profile measurements above mid-latitude North America during the IONS/ICARTT Campaign: Evidence for stratospheric input, *Eos Trans. AGU,* **86**(52), Fall Meet. Suppl., Abstract A33F, 2005.

Teitelbaum, H., J. Ovarlez, H. Kelder, and F. Lott, Some observations of gravity-wave-induced structure in ozone and water vapour during EASOE, *Geophys. Res. Lett.*, **21,** 1483-1486, 1994.

Teitelbaum, H., M. Moustaoui, J. Ovarlez, and H. Kelder, The role of atmospheric waves in the laminated structure of ozone profiles at high latitudes, *Tellus,* **48A,** 442-455, 1996. et al., 1996.

Thompson, A. M., L. C. Sparling, Y. Kondo, B. E. Anderson, G. L. Gregory, and G. W. Sachse, Perspectives on NO, NO_y and fine aerosol sources and variability during SONEX, *Geophys. Res. Lett.*, **26**, 3073-3076, 1999.

Thompson, A. M., B. G. Doddridge, J. C. Witte, R. D. Hudson, W. T. Luke, J. E. Johnson, B. J. Johnson, S. J. Oltmans, R. Weller, A tropical Atlantic ozone paradox: Shipboard and satellite views of a tropospheric ozone maximum and wave-one in January-February 1999, *Geophys. Res. Lett.*, **27,** 3317-3320, 2000.

Thompson, A. M., et al., IONS-2004 (INTEX Ozonesonde Network Study) ozone budgets: Experimental determination and comparison with climatology, *Eos Trans. AGU*, **86**(52), Fall Meet. Suppl., Abstract A11B Fall AGU, 2005.

Thouret, V., A. Marenco, J. A. Logan, P. Nédélec, C. Grouhel, Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations, J. Geophys. Res., 103(D19), 25695-25720, 10.1029/98JD02243, 1998.

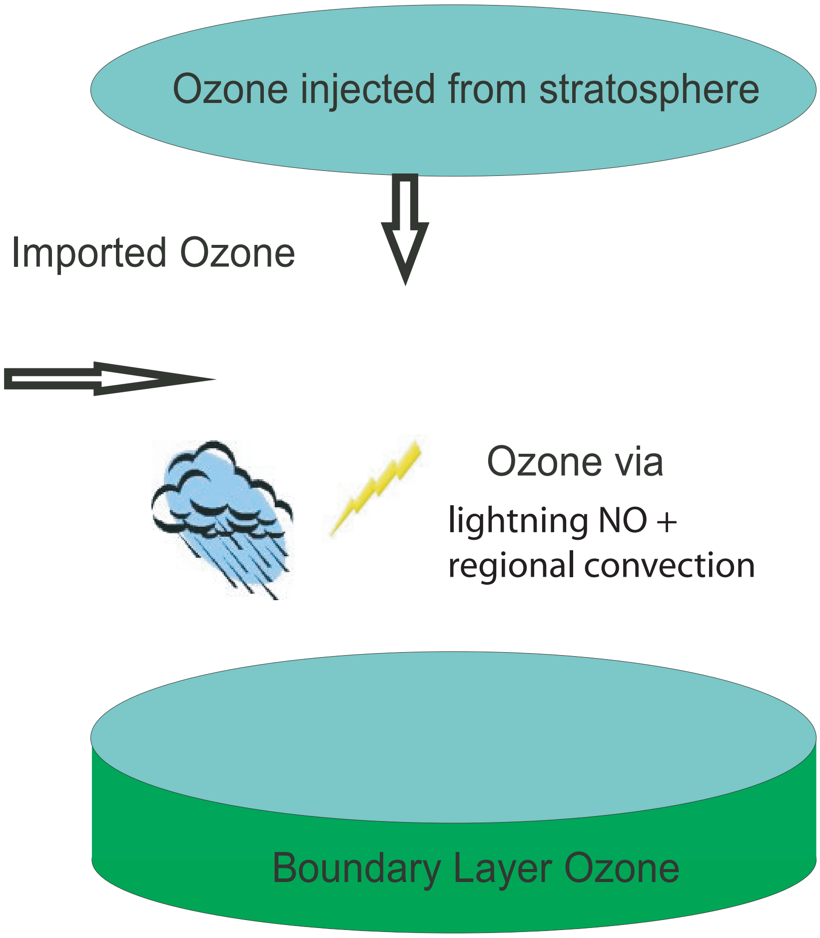


FIG. 1

Figure 1. Schematic of contributions to the ozone column budget. Stratospheric O_3 may contribute at any level in the free troposphere. Boundary layer ozone represents "local" pollution. Interactions of local O_3 precursors with regional convection and lightning define a third O_3 source. The remainder is assumed to be a net advection source.

FIG. 2

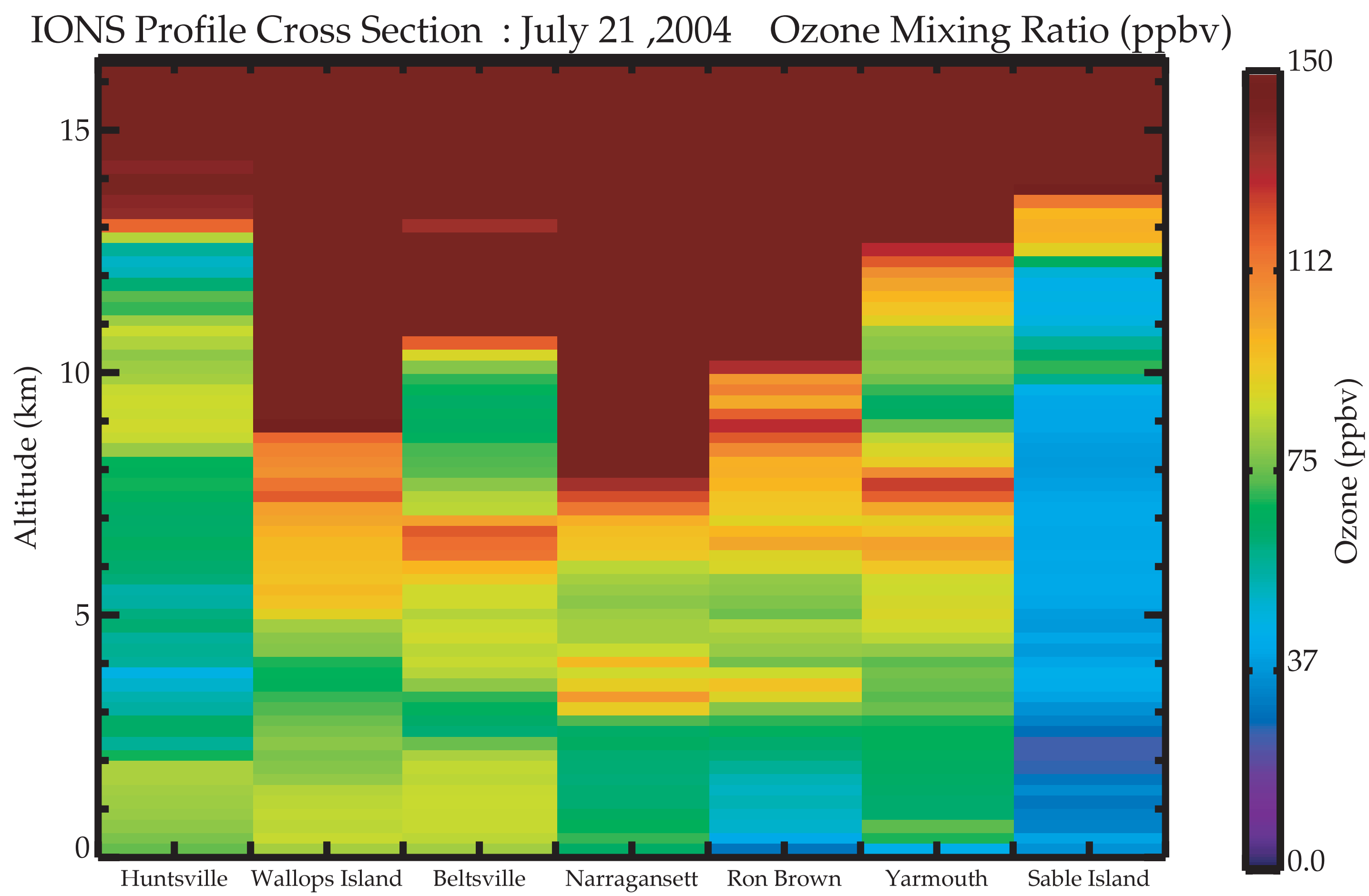


Figure 2. Cross-section of O_3 mixing ratios below 17 km on 21 July 2004 from IONS soundings with sites arranged by increasing latitude. The corresponding longitudes order sites from south central to northeast US-maritime Canada. The four New England-Maritime Canada stations (Narragansett, *R/V R H Brown*, Yarmouth and Sable Island) display a progression of tropopause height (the red-brown transition in most cases) that increases with increasing latitude and eastward-increasing longitude (coordinates in Table 1).

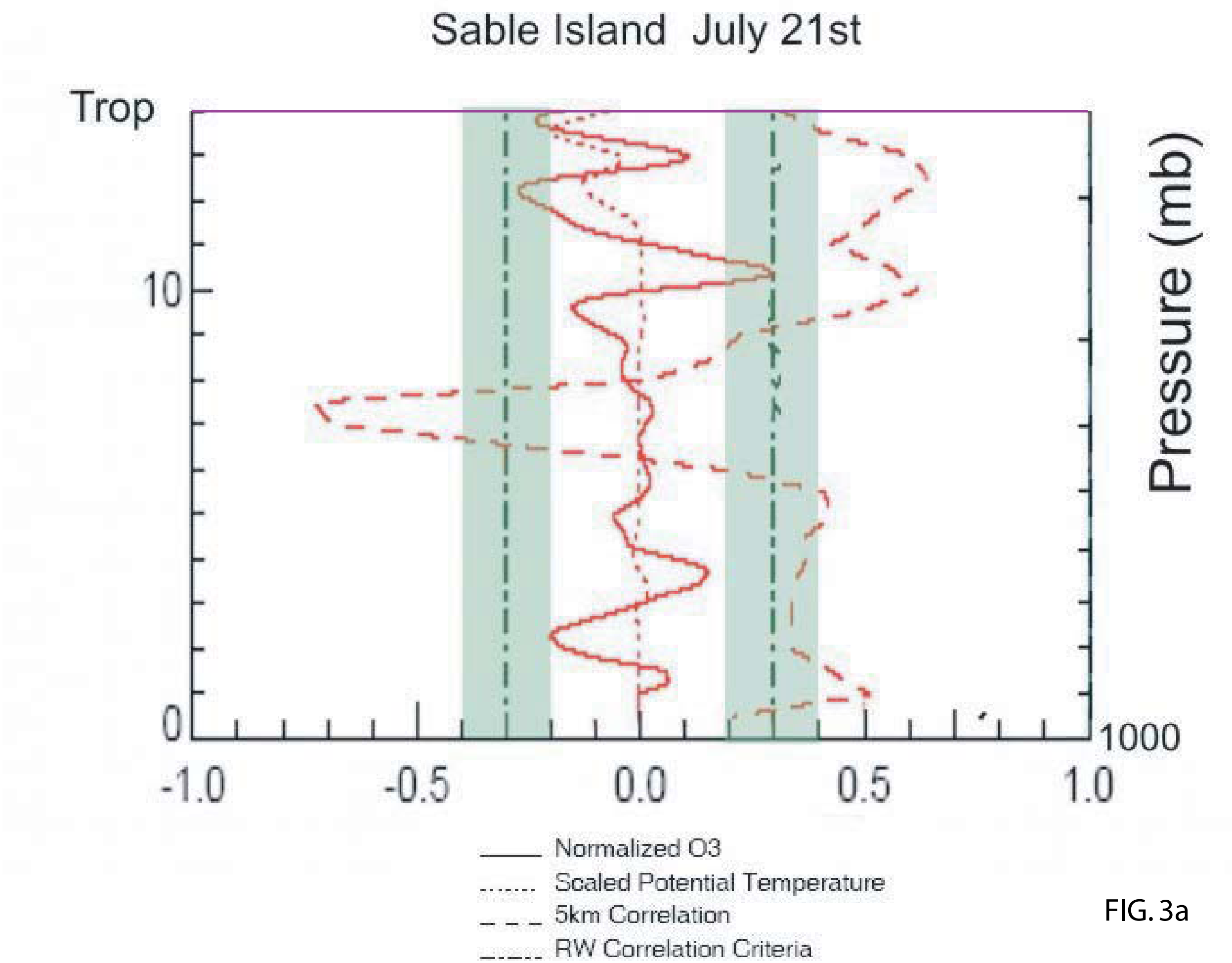


FIG. 3a

Figure 3. Elements of PT analysis for three ozone profiles shown in Figure 2: (a) Sable Island; (b) Narragansett, *R/V R H Brown*. Ozone laminae (normalized relative to a running mean within the profile) are given by solid lines. Dotted lines are normalized potential temperature (θ) laminae from P-T-U measurements of the radiosonde flown with the ozonesonde. Dashed lines signify correlations between the O_3 and θ gradients. Low correlations, within the green-shaded ranges, are attributed to Rossby-waves (RW) and potential introduction of stratospheric air into the troposphere. The corresponding O_3 layer amounts (RW O_3) are integrated and added together to determine the total RW O_3 for the sounding. "Minimum" RW O_3 in Table 2 is based on lower-limit RW designations and O_3 column amounts (and fractions) that correspond to dashed lines falling between -0.2 and 0.2 (bounded by green shading). Upper limits for RW O_3 (Table 2 "maximum") based on -0.4 to 0.4 (outer bounds of shading) as low-correlation limits. The PT method (Figure 1 in Pierce and Grant, 1998) assigns Gravity Wave (GW) character to high correlations (> 0.7) between O_3 and θ gradients. Where GW layers are associated with air parcels exposed to convection and lightning during INTEX-NA (e.g, EL images at <croc.gsfc.nasa.gov/intex>) the O_3 may originate from regional convection and lightning [Thompson et al., 2005]. After summing BL, ST and convectively influenced O_3, a residual tropospheric O_3 is assigned to transport. See supplementary Material for RW, GW, and BL O_3 for Narragansett and Wallops. During IONS, advected O_3 came from Asia as well as Alaskan and Canadian wildfires, the latter prominent in summer 2004 [Pfister et al., 2005a,b; Morris et al., 2006].

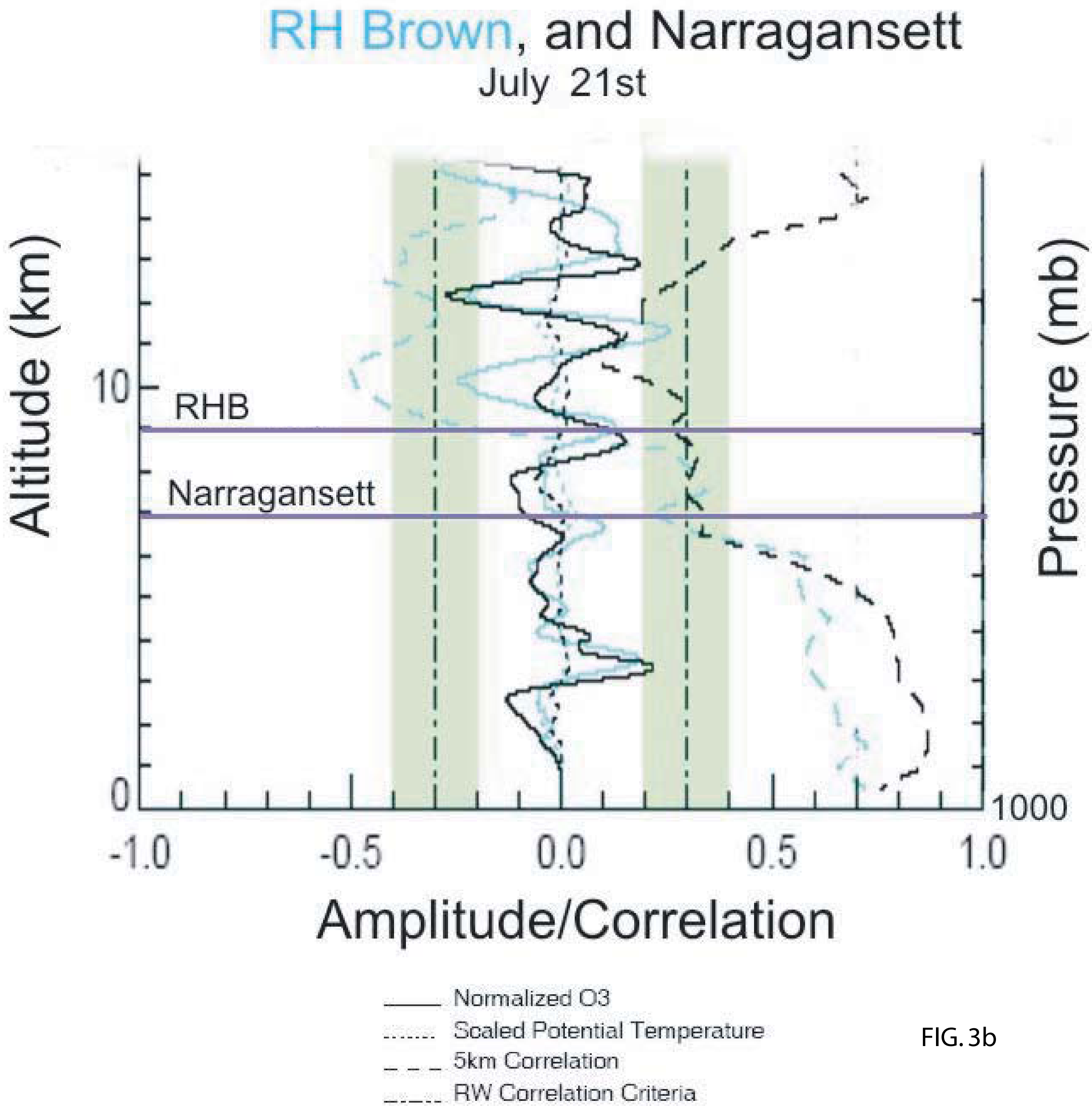
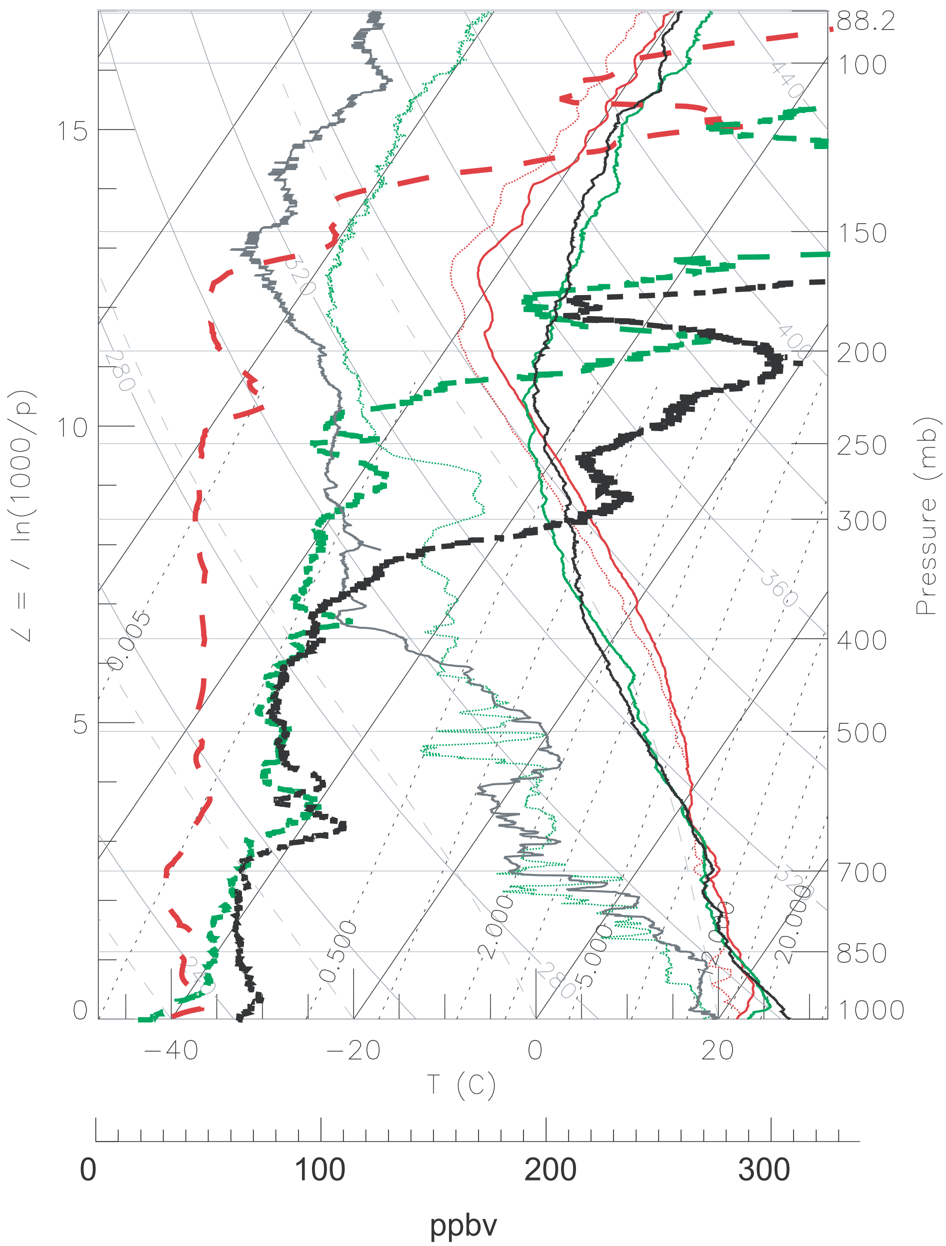


FIG. 3b

Figure 3. Elements of PT analysis for three ozone profiles shown in Figure 2: (a) Sable Island; (b) Narragansett, *R/V R H Brown*. Ozone laminae (normalized relative to a running mean within the profile) are given by solid lines. Dotted lines are normalized potential temperature (θ) laminae from P-T-U measurements of the radiosonde flown with the ozonesonde. Dashed lines signify correlations between the O_3 and θ gradients. Low correlations, within the green-shaded ranges, are attributed to Rossby-waves (RW) and potential introduction of stratospheric air into the troposphere. The corresponding O_3 layer amounts (RW O_3) are integrated and added together to determine the total RW O_3 for the sounding. "Minimum" RW O_3 in Table 2 is based on lower-limit RW designations and O_3 column amounts (and fractions) that correspond to dashed lines falling between -0.2 and 0.2 (bounded by green shading). Upper limits for RW O_3 (Table 2 "maximum") based on -0.4 to 0.4 (outer bounds of shading) as low-correlation limits. The PT method (Figure 1 in Pierce and Grant, 1998) assigns Gravity Wave (GW) character to high correlations (> 0.7) between O_3 and θ gradients. Where GW layers are associated with air parcels exposed to convection and lightning during INTEX-NA (e.g, EL images at <croc.gsfc.nasa.gov/intex>) the O_3 may originate from regional convection and lightning [Thompson et al., 2005]. After summing BL, ST and convectively influenced O_3, a residual tropospheric O_3 is assigned to transport. See supplementary Material for RW, GW, and BL O_3 for Narragansett and Wallops. During IONS, advected O_3 came from Asia as well as Alaskan and Canadian wildfires, the latter prominent in summer 2004 [Pfister et al., 2005a,b; Morris et al., 2006].



Sable, RH Brown, and Narragansett

July 21st, 2004

FIG. 4

Figure 4. Three IONS O_3 profiles for 21 July 2004 (dashed lines; corresponding normalized laminae appear in Figure 3) with coincident temperature (T, bolder shade) and dewpoint temperature (DPT, paler shade) from the radiosonde launched with the ozonesonde.

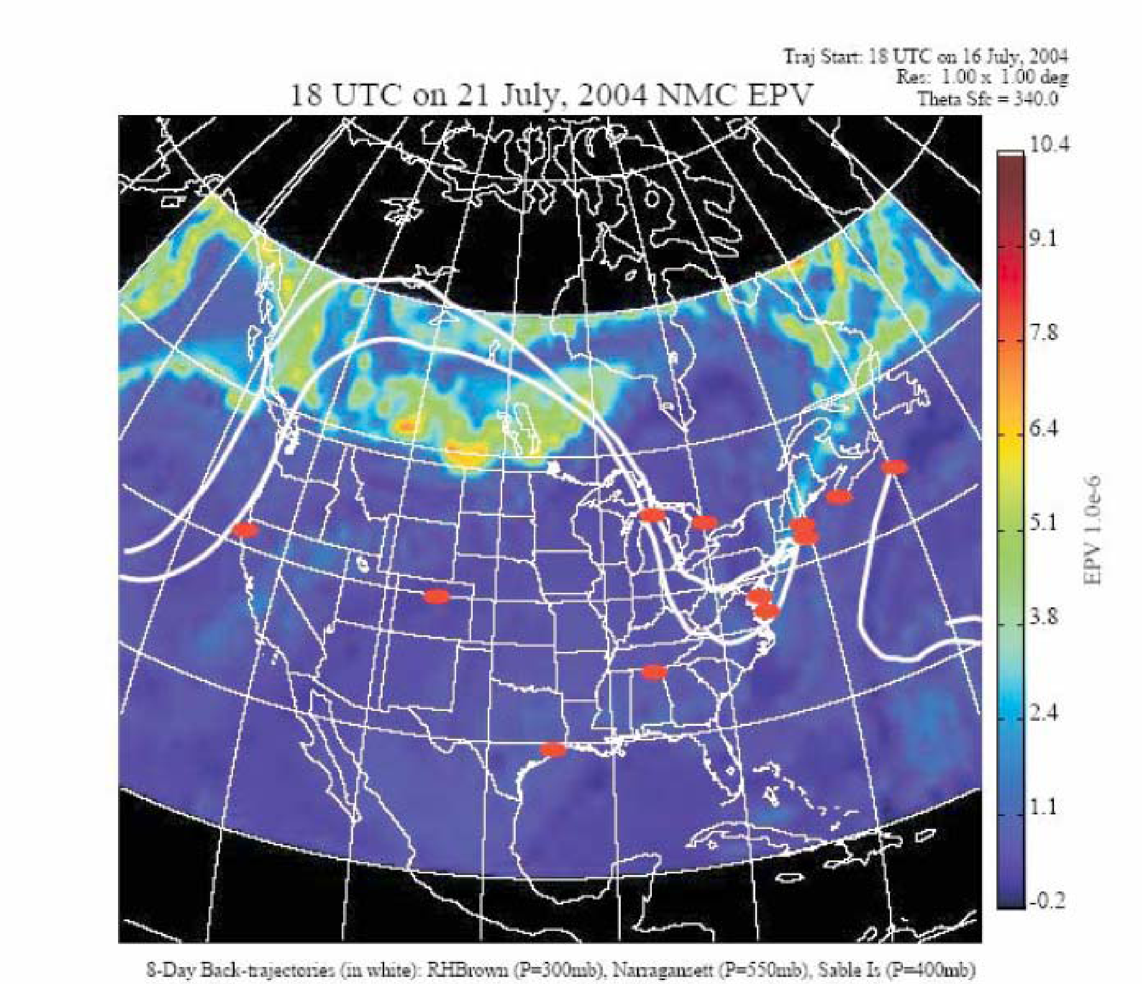


FIG. 5

Figure 5. IONS sites (red dots) with origins of 8-day back-trajectories (in white) from sites shown in Figures 3 and 4 (Narragansett, *R/V R H Brown*, Sable Island) superimposed on trajectory-mapped epv for 21 July 2004. Levels chosen for the trajectory initialization correspond to RW laminae (Figure 3). Pv and meteorological fields used in trajectories (initialized at 1°x1° resolution) are taken from the Goddard Assimilation Model (GEOS 4 version; Bloom et al., 2005). Trajectory mapping (as depicted in the map) displays the 5-day cumulative impact of pv in air parcels arriving each grid point (340K here, ~ 10 km) in contrast to an instantaneous pv value at the location.

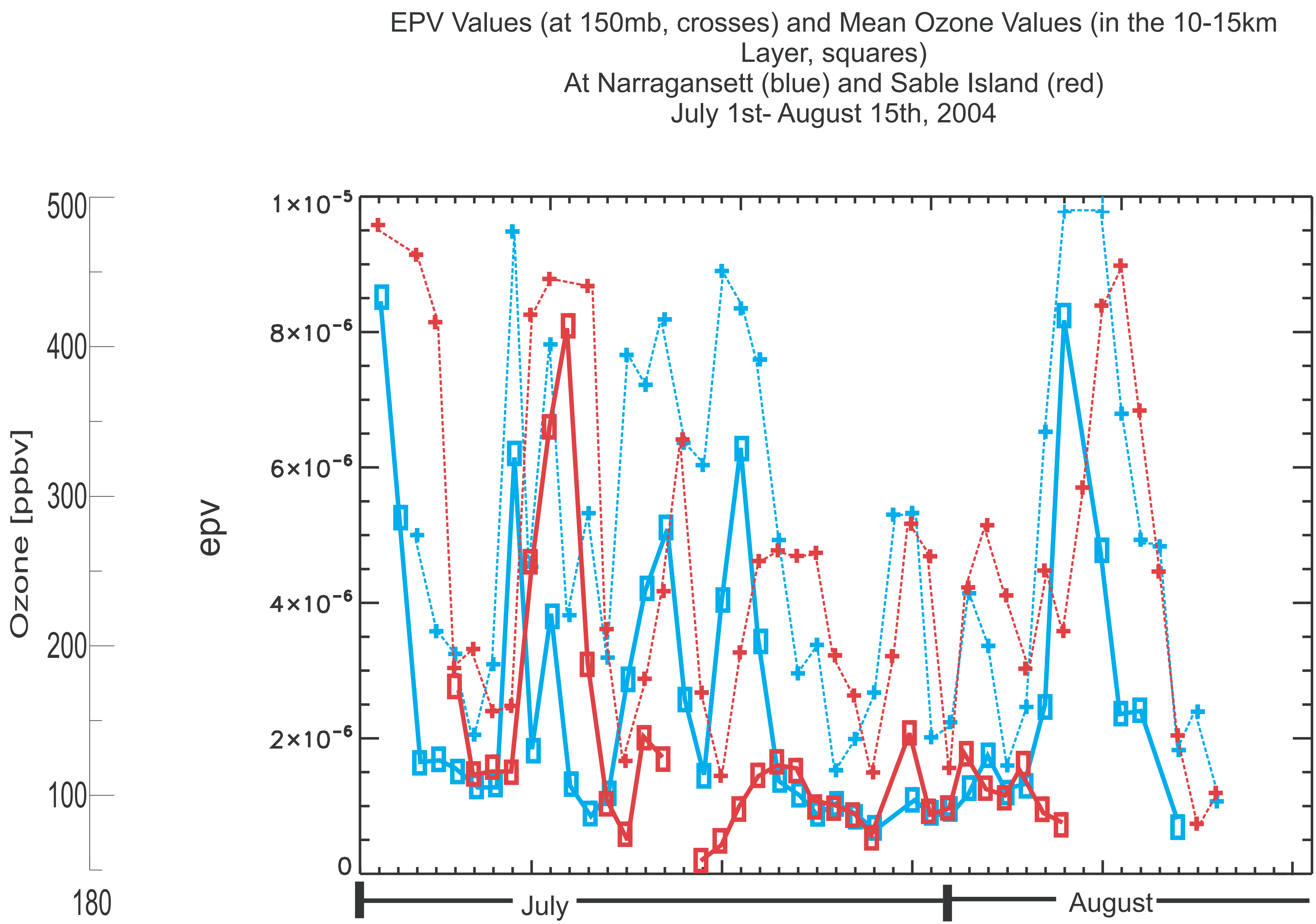


FIG. 6

Figure 6. (a) Daily pv (interpolated at 150 hPa, ~13 km, from GEOS 4 analyses) from 1 July to 15 August 2004 (+) with O_3 mixing ratio averaged between 10 and 15 km (□) at Narragansett and Sable Island. The *R/V R H Brown* and Yarmouth pv and O_3 are similar to Narragansett. (b) Scatter-plot of ozone and pv (150 hPa) over six IONS sites.

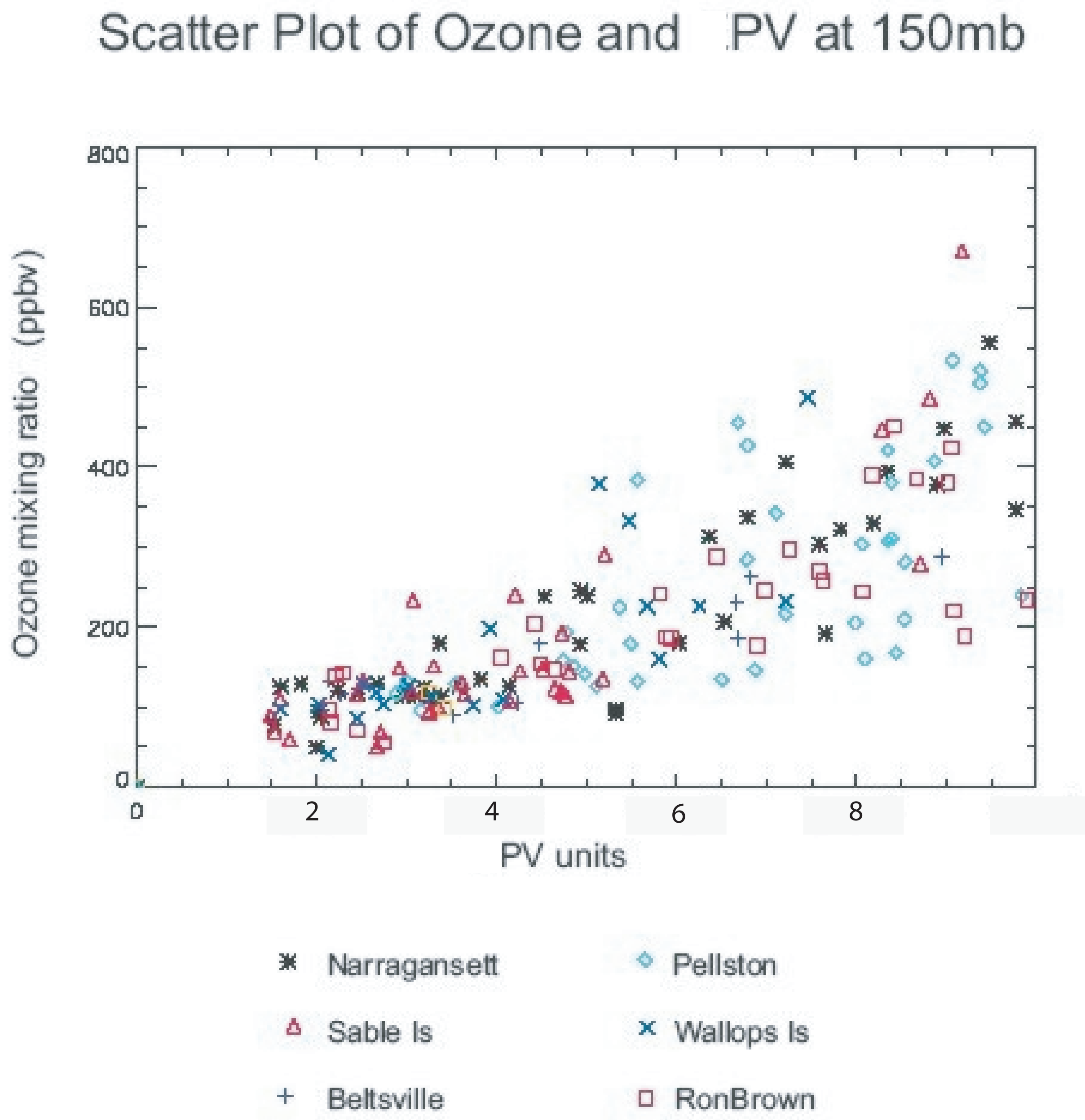


Figure 6. (a) Daily pv (interpolated at 150 hPa, ~13 km, from GEOS 4 analyses) from 1 July to 15 August 2004 (+) with O_3 mixing ratio averaged between 10 and 15 km (□) at Narragansett and Sable Island. The *R/V R H Brown* and Yarmouth pv and O_3 are similar to Narragansett. (b) Scatter-plot of ozone and pv (150 hPa) over six IONS sites.

ST, RW, and 10-15km Ozone as a Fraction of the Total Column

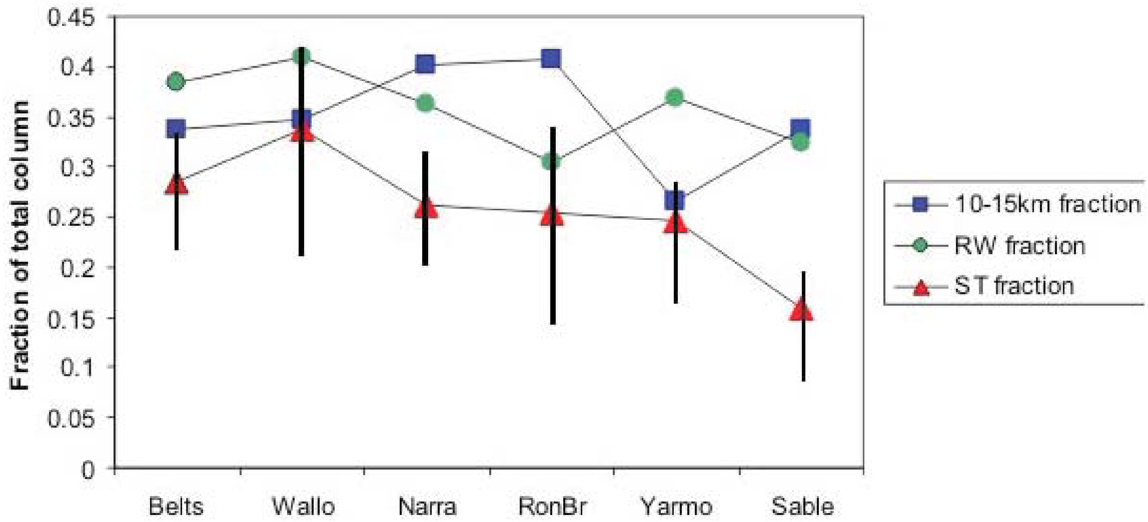


FIG. 7a

Figure 7 (a) Mean RW and ST (fraction of stratospherically influenced O_3 below tropopause, Table 3) at designated IONS sites and the mean fraction of O_3 within the layer 10-15 km compared to the total surface-to-15 km O_3 column. RW and ST O_3 amounts are based on averages of minimum and maximum RW-OTP (Table 3). (b) Fractions of O_3 within 9-11 km layer relative to O_3 column from surface to 11 km for IONS sites and MOZAIC LTO profiles at four NENA airports during JJA 2004.

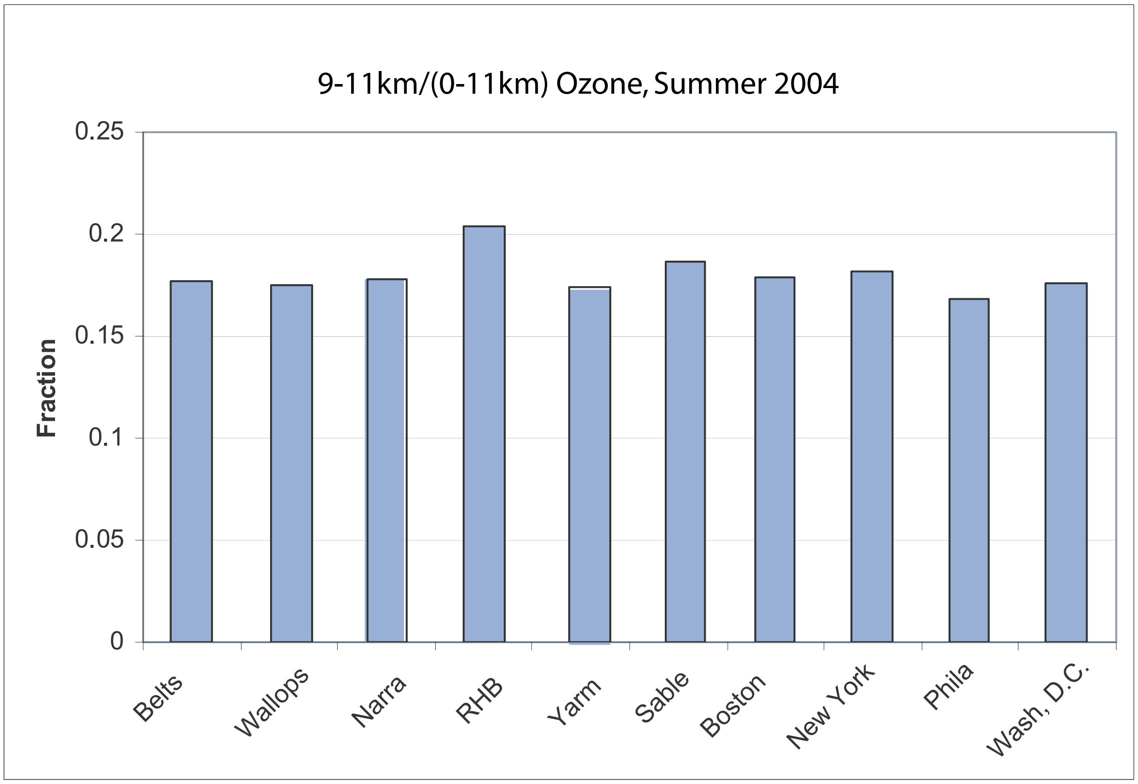


FIG. 7b

Figure 7 (a) Mean RW and ST (fraction of stratospherically influenced O_3 below tropopause, Table 3) at designated IONS sites and the mean fraction of O_3 within the layer 10-15 km compared to the total surface-to-15 km O_3 column. RW and ST O_3 amounts are based on averages of minimum and maximum RW-OTP (Table 3). (b) Fractions of O_3 within 9-11 km layer relative to O_3 column from surface to 11 km for IONS sites and MOZAIC LTO profiles at four NENA airports during JJA 2004.

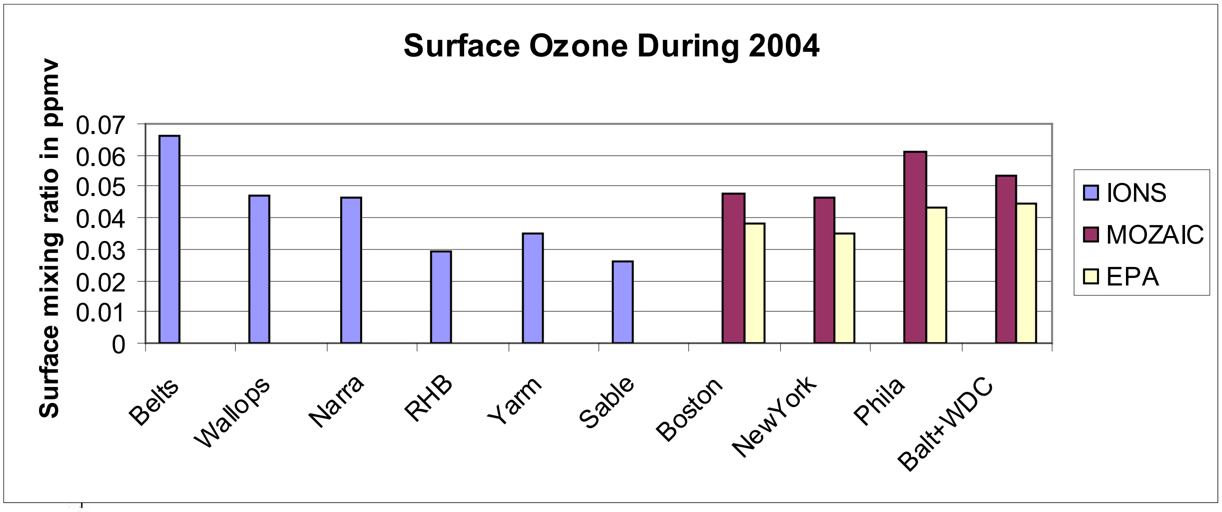


FIG. 8

Figure 8. Near-surface O_3 mixing ratio (surface to 100 m) at six IONS sites, averaged during 1 July to 15 August 2004, and mean JJA 2004 surface O_3 from four MOZAIC airports (Boston, New York, Philadelphia, Washington).

Wallops Ozone

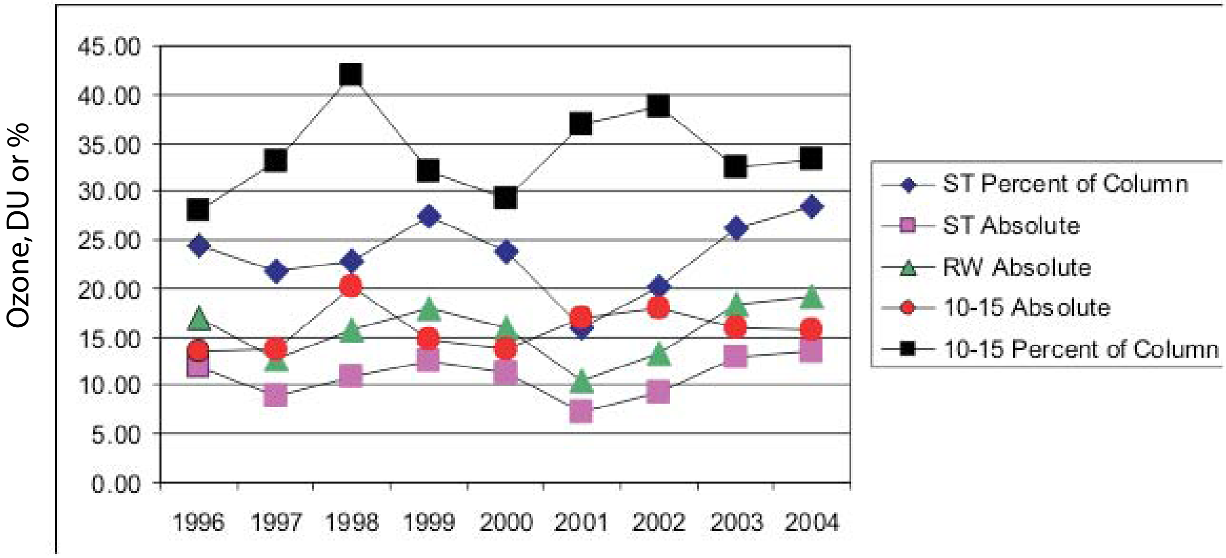


FIG. 9a

Figure 9. (a) Mean ST O_3 (DU and fraction) from JJA 1996-2004 at Wallops evaluated by the PT method with mean 10-15 km O_3 amounts from the same soundings; RW fraction also displayed; (b) 9-11 km O_3 amount and fractions from JJA soundings at Wallops and for JJA MOZAIC profiles at New York, Washington, Boston and Philadelphia.

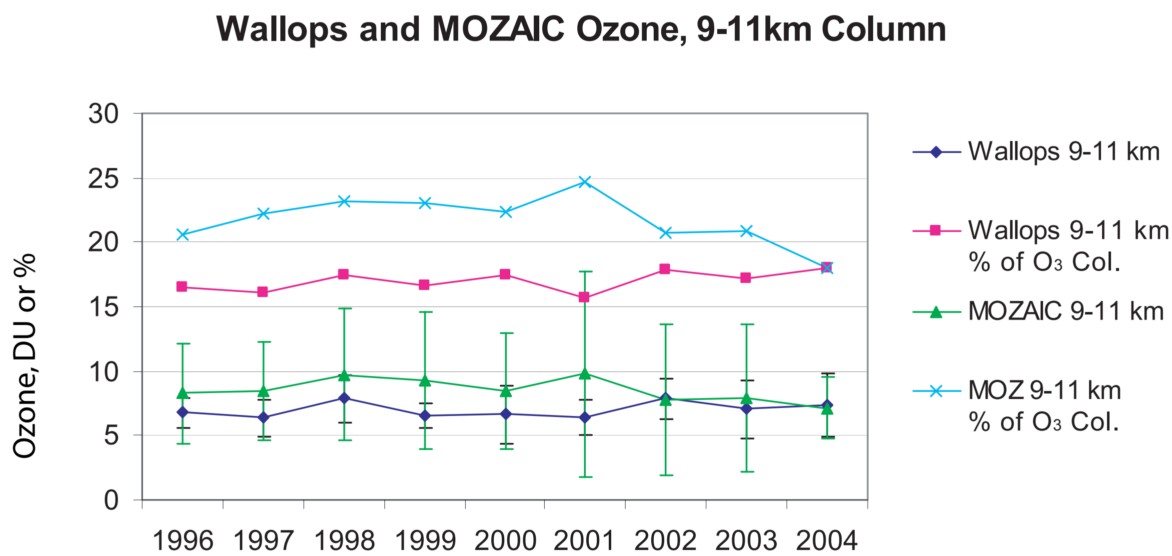


FIG. 9b

Figure 9. (a) Mean ST O_3 (DU and fraction) from JJA 1996-2004 at Wallops evaluated by the PT method with mean 10-15 km O_3 amounts from the same soundings; RW fraction also displayed; (b) 9-11 km O_3 amount and fractions from JJA soundings at Wallops and for JJA MOZAIC profiles at New York, Washington, Boston and Philadelphia.

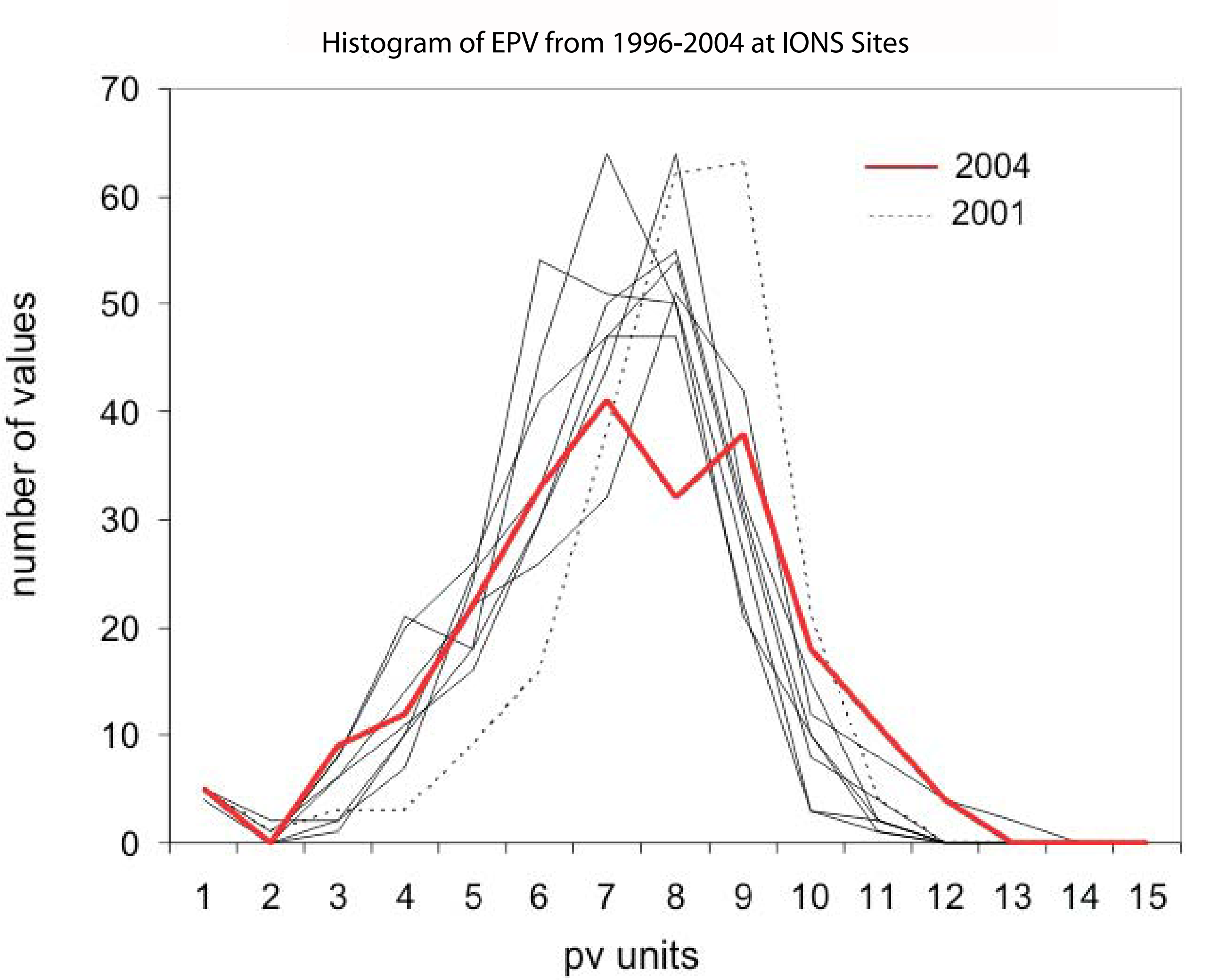


FIG. 10

Figure 10. Histograms of pv for July 1 through 15 August for years 1996-2004 averaged over locations corresponding to Pellston, Beltsville and Wallops (same grid point in the meteorological analysis); Narragansett-Yarmouth- *R/V R H Brown* (same value in the grid); Sable Island. Statistical tests (e.g. Kruskal-Wallace) to determine whether any year(s) were distinct showed that 2004 (red) was not; only 2001 (dashed) was unique.

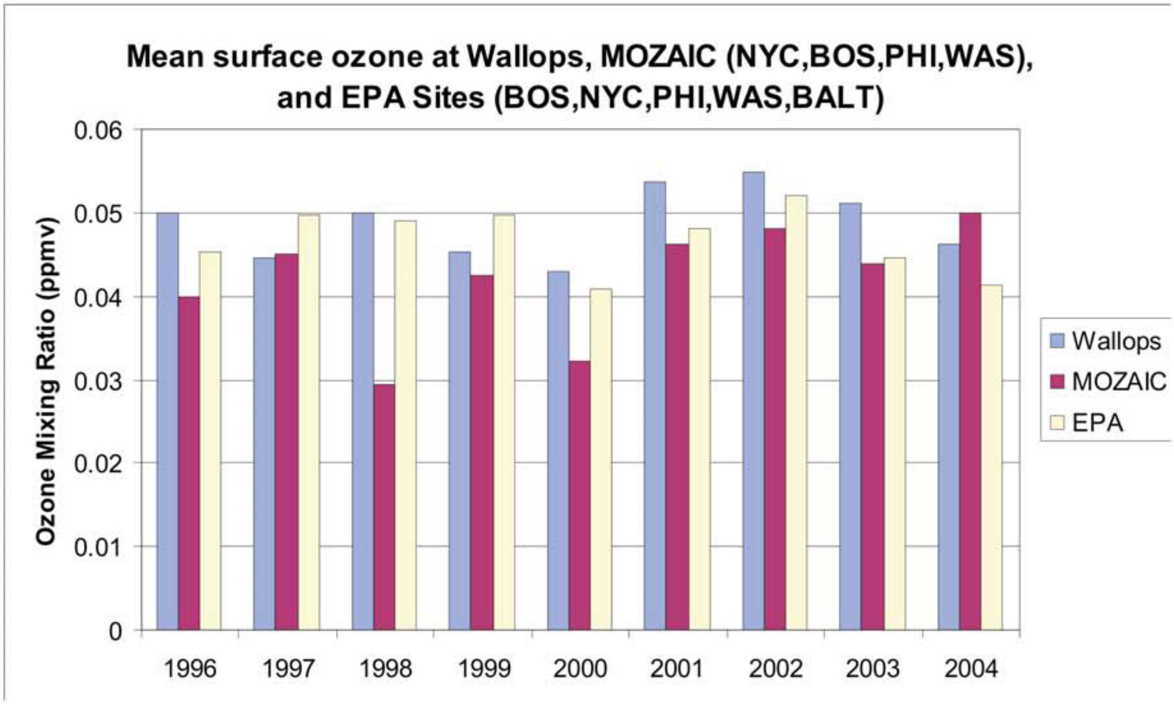


FIG. 11

Figure 11. Surface O_3 (mean from surface to 0.1 km) for 1996-2004 (during JJA) from Wallops soundings (blue) and average from MOZAIC LTO profiles at four NENA airports (red). Same profiles used as in Figure 9. Also displayed are daytime (0900-2100 hr Local, averaged, in yellow) surface O_3 readings from stations around five NENA cities (New York, Washington, Baltimore, Boston, Philadelphia). Supplementary Material gives city-by-city annual mean and median EPA O_3 values.

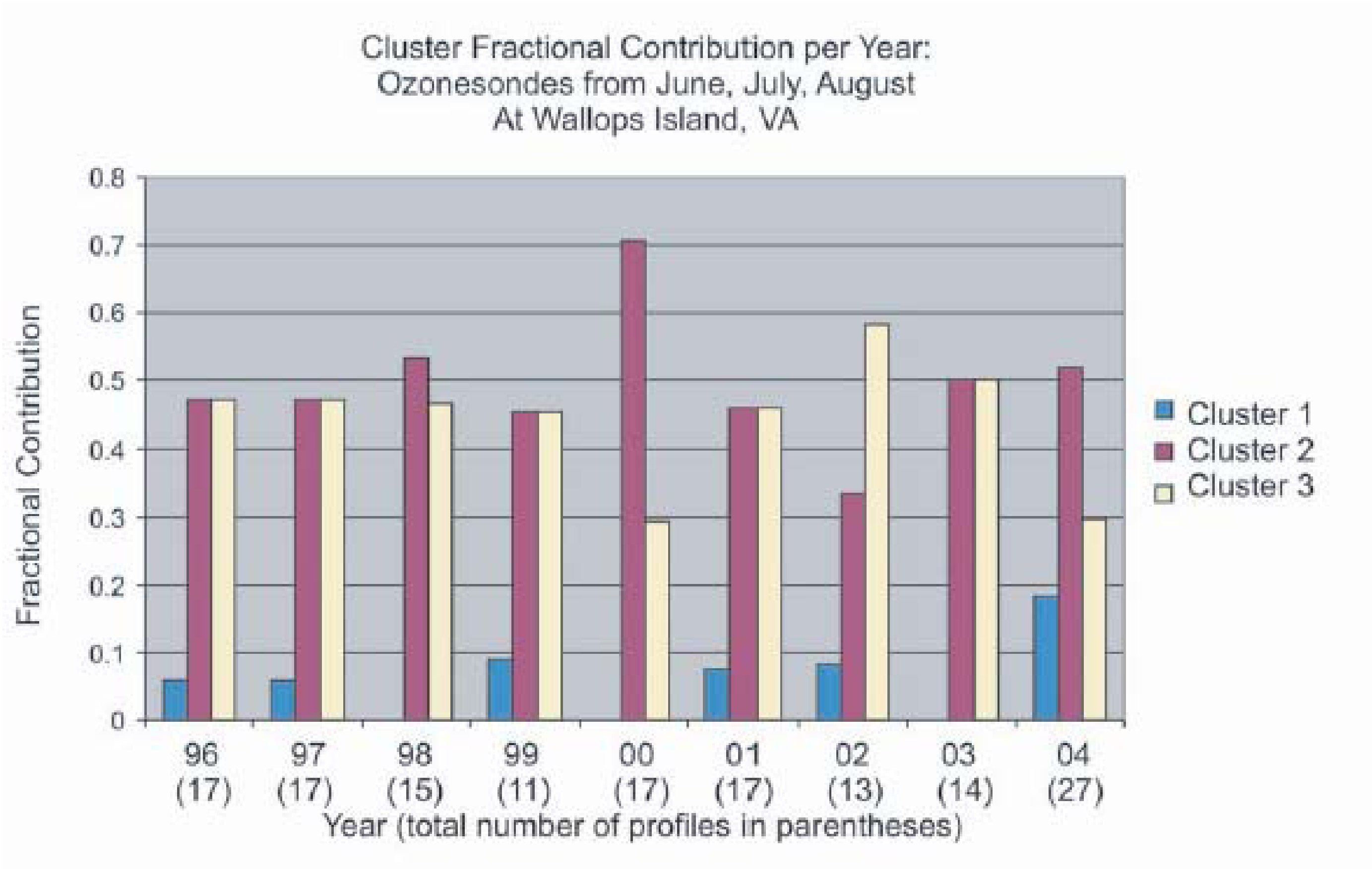
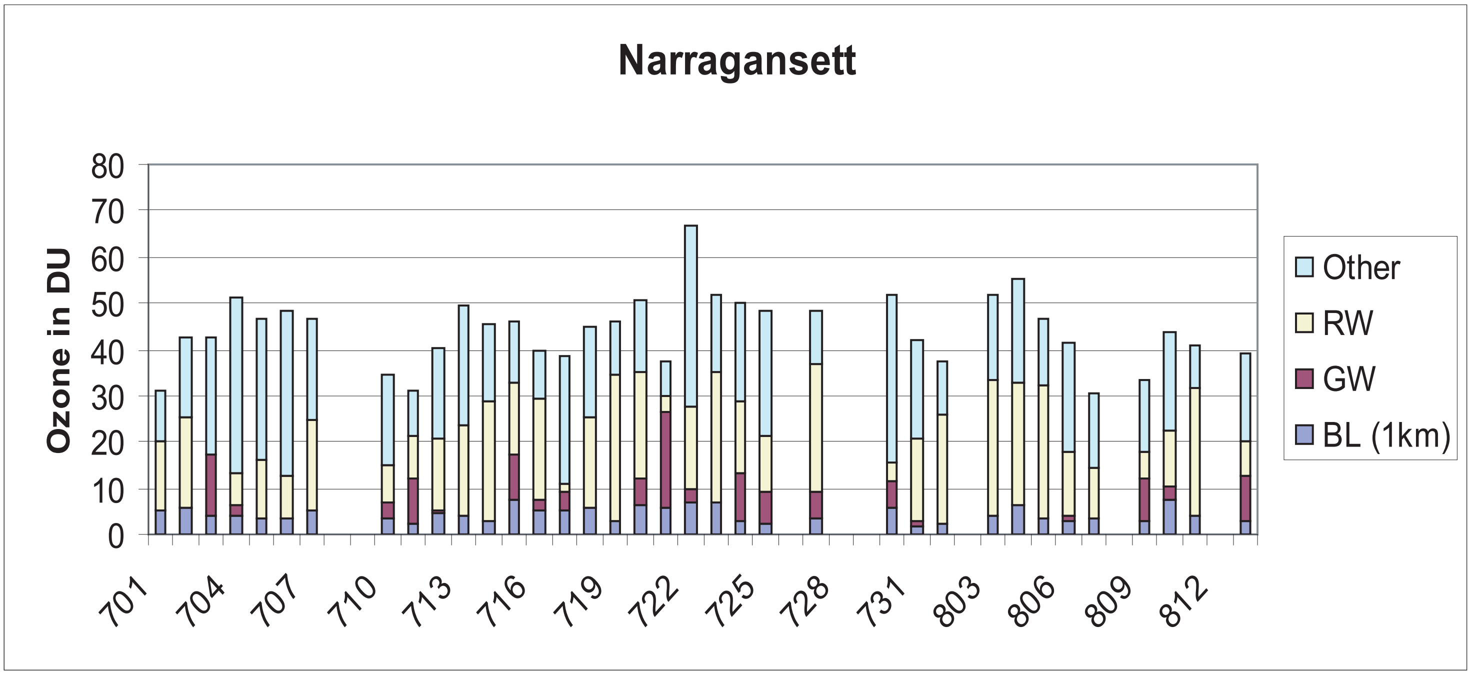
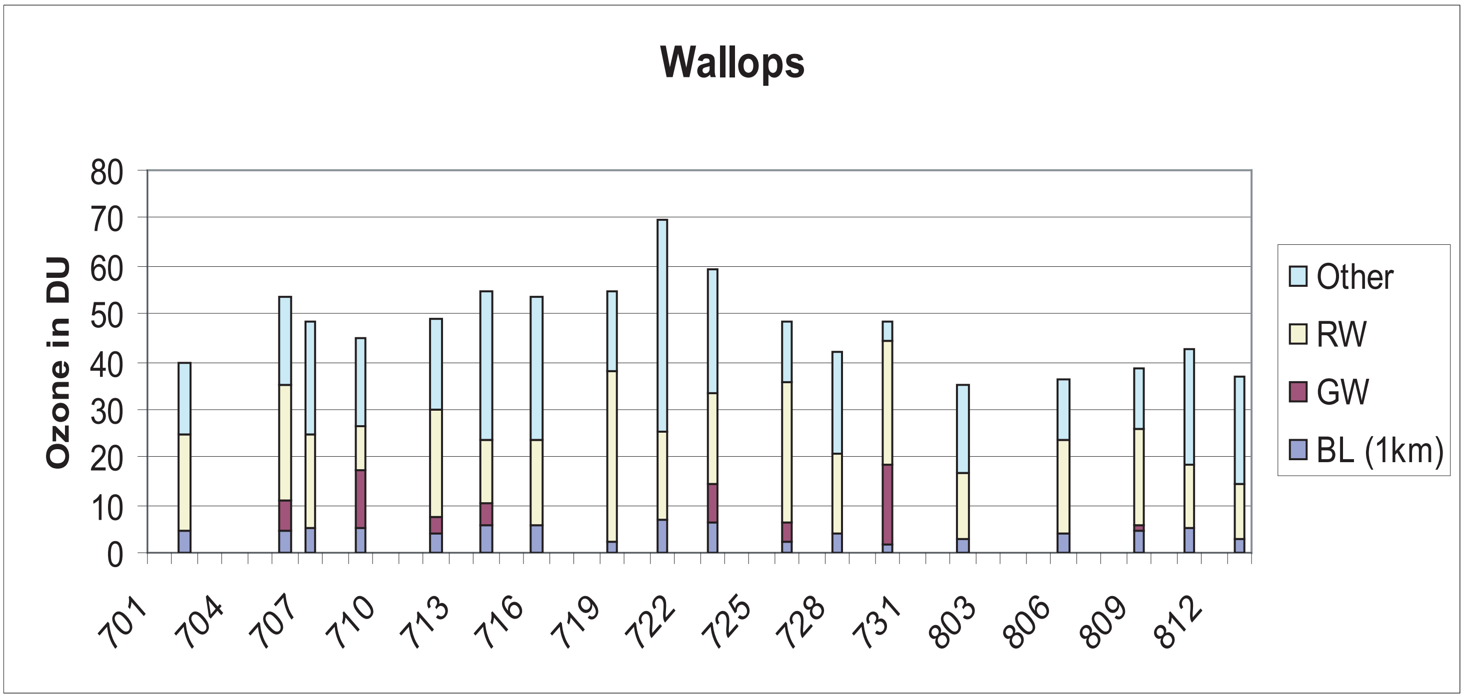


FIG. 12

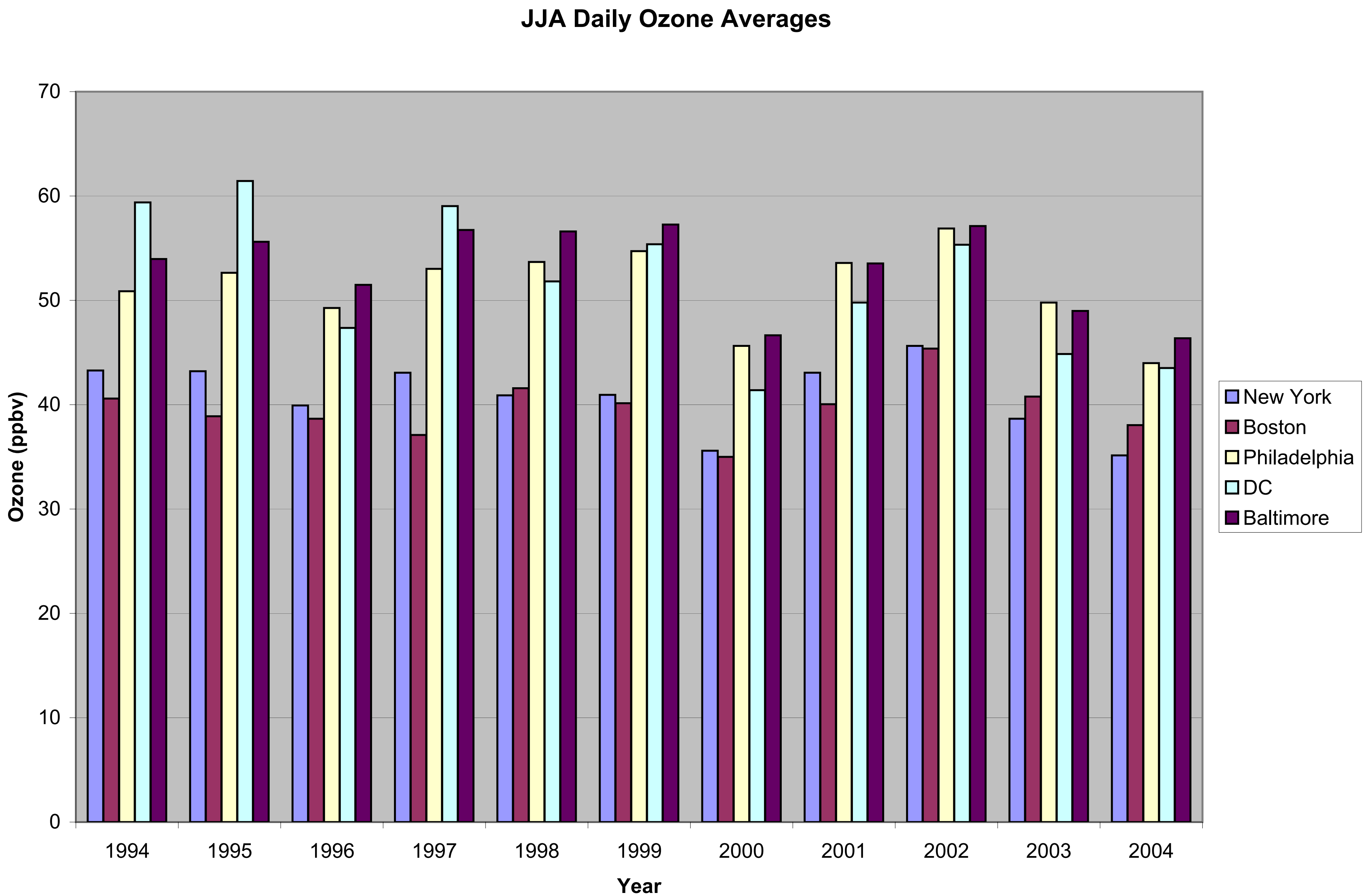
Figure 12. Histogram of the fractions of three statistically classified types of JJA O_3 profiles (from 0-10 km) for 1996-2004. The number of soundings for each JJA is at bottom. Profiles assigned to Type #1 represent a low "background" marine-type profile in which O_3 declines monotonically above the surface. Supplementary Material displays clusters. Classes 2 and 3 are discriminated by degree of O_3 pollution in the lower troposphere, with Class 3 having greatest ozone below 600 hPa. Class 2 fraction for 2004 at Wallops is not exceptional but Class 1 profiles are the highest fraction in 1996-2004. Class 3 fraction is a low value matched in only one other year. The years 2002 and 2004 represent an opposite distribution. Violations of the National Ambient Air Quality Standard for ozone numbered 18 in Maryland in 2002; only one violation occurred in 2004. Similar statistics applied to New York, Rhode Island and Massachusetts.

Supplementary material for Figure 3.

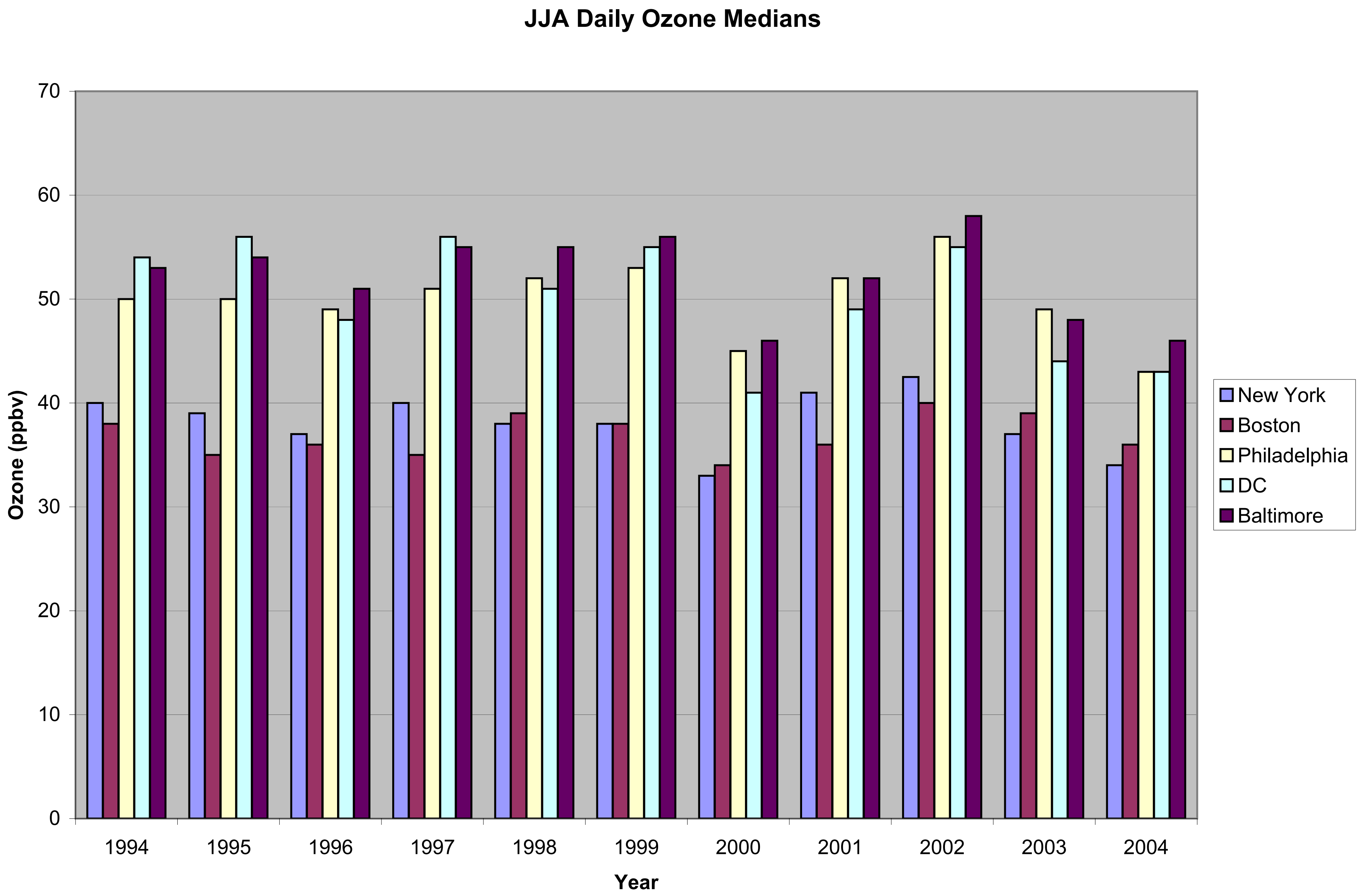




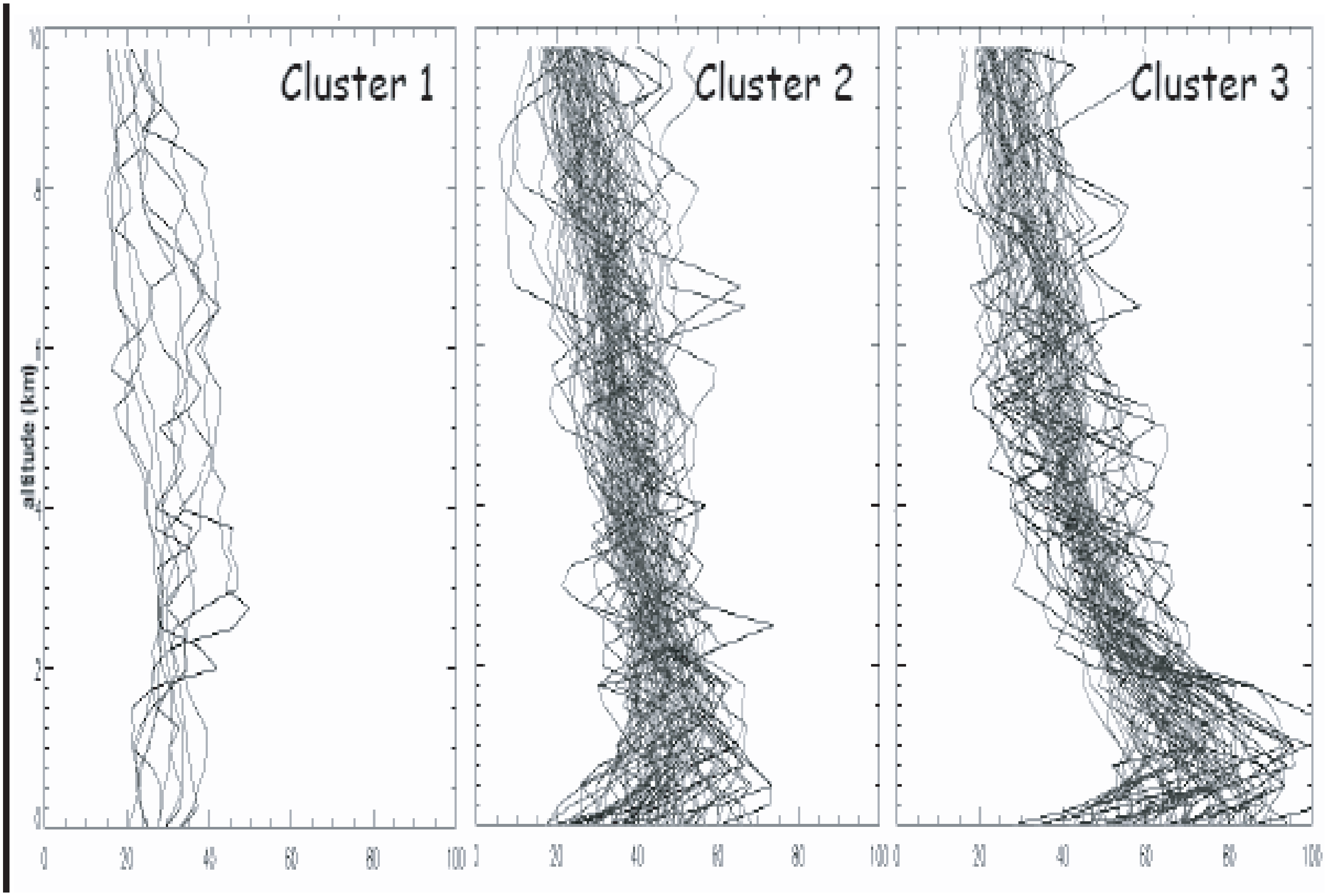
Supplementary material for Figure 11



Supplementary material for Figure 11



Supplementary Material for Figure 12



ozone in nb